

澳门环境与城市发展科学研究

INVESTIGAÇÃO CIENTÍFICA SOBRE O AMBIENTE E DESENVOLVIMENTO
URBANO EM MACAU

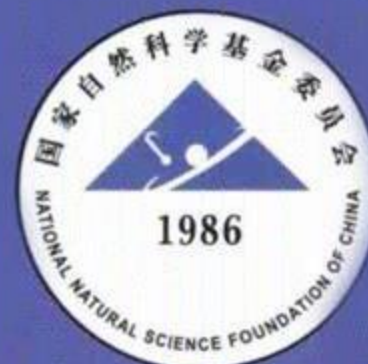
王志石 陈吉宁 杜鹏飞



澳门大学



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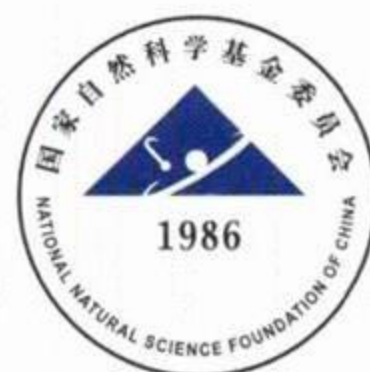
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2002

澳門環境與城市發展科學研究

主編

王志石 陳吉寧 杜鵬飛

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PREFACE

The joint research program on Macau environment and city development has been carrying out since 1994, jointly sponsored by National Natural Science Foundation and University of Macau as well as Macau Foundation. Under such a cooperation framework, the professors in Department of Environmental Science and Engineering, Tsinghua University, Beijing have actively participated in the collaborative researches for years and contributed to the research program productively and constructively. Together with the professors in Faculty of Science and Technology, University of Macau, Macau, they have studied the Macau urban air and noise pollutions and the coastal water environment surrounding Macau by field monitoring, computer modeling, as well as remote sensing technique. Part of the updated research achievements was published in this book, together with research contributions by the other joint research program partners in Department of Hydraulic and Hydropower Engineering, Tsinghua University, and in Guangzhou Research Institute of Geochemistry, Chinese Science Academy, and in Environmental Science Research Center, Beijing University. The local research teams of Macau have also contributed their research achievements to publication of this book. We are sincerely grateful for all these contributors.

Zhishi Wang, Jining Chen, Pengfei Du

前 言

自 1994 年以来, 在国家自然科学基金委员会, 澳门大学及澳门基金会的联合资助下, 清华大学环境科学与工程系与澳门大学科技学院在澳门环境与城市发展领域进行了卓有成效的合作研究, 共同研究了澳门市区空气和噪音污染, 遥感在澳门水域环境研究的应用, 南湾湖生态环境评价等。清华大学水利水电工程系教育部泥沙重点实验室, 中科院广州地球化学研究所以及北京大学环境科学研究中心等也参与了这项合作研究计划, 并在澳门河口污染物迁移数学模型, 澳门大气和水环境中的有害有机物污染监测与评价, 澳门街区风洞试验等方面做出了很有价值的贡献。他们研究的最新成果都收入此论文集之中。澳门大学科技学院教授们的有关研究成果也一并收入论文集中, 包括澳门空气质量预测预报模型, 室内空气质量控制技术。在此, 我们向所有作者表示衷心的感谢。

王志石, 陈吉宁, 杜鹏飞

2002 年 4 月

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Fate and Pathways of Organic and Inorganic Toxicant Pollution in Macau Coastal Waters

Zhishi Wang

Faculty of Science and Technology, University of Macau, Macau

ABSTRACT: Toxicant pollution usually involves persistent organic pollution (PAHs, PCBs, organochlorine pesticides) and heavy metals (Pb, Cd, Cr, etc) in water and sediment as well as biota phases. A long term field monitoring research has been made since 1994 regarding to the toxicant pollution in Macau coastal waters, aiming at finding out the pollution sources of the specific pollutants such as PAHs and tracing back to the pollution histories such as DDTs residue variation along the sediment column samples (1 m depth) as well as identifying high risky pollution zones of the coastal waters (such as Porto Interior or inner port). However, it has been found that solely based on the field monitoring data, the above-listed tasks can only be partially completed, particularly for identification of fate and pathways of the toxicant pollution in the coastal waters. Therefore, it is vitally needed to establish the conceptual models to insight the complex pollution processes occurring in the coastal waters, especially these processes such as volatile organic species exchange between atmosphere and water phases, adsorption and absorption of inorganic and organic compounds on suspended particulate matter, particle settling and pollutants vertical transport, and mass transfer at water/sediment interfaces, as well as tide flow patterns and long distance pollution transport in the coastal waters.

Macau coastal waters are characterized by high turbidity with high suspended solid contents in water column and active mass transfer at water/sediment interfaces. For fate and pathways of organic and inorganic toxic pollution in the coastal waters, it is believed that the pollutant adsorption and absorption on suspended solid surfaces and exchange between water/sediment phases are important while the atmosphere/water exchange is not, because of nonvolatile nature of the persistent organic pollutants. The study of conceptual modeling for the pollution fate and pathways was focused on the water-particulates interaction and exchange processes, particularly for water-solid phase partitioning of persistent organic pollution (e.g., PAHs) in water column and in surface sediments as well as for adsorption and remobilization of heavy metals in the coastal water sediments.

KEYWORDS: water-sediment phase partitioning, adsorption and absorption, neutral no polar organic compounds, heavy metals

Fate and Pathways of Organic and Inorganic Toxicant Pollution in Macau Coastal Waters

Usually an estuary converges a large quantity of runoff from a wide basin such as the Pearl River delta, leading to transport of continent pollutants including anthropogenic inorganic and organic toxicants to the estuary. Urban runoff outfalls from a metropolitan coastal city like Macau also exert adverse impacts on the estuary such as nutrient overloads and persistent toxicant accumulation. The acid rain widely occur in the Pearl River delta including Macao and its neighboring areas, which contributes to nutrient and toxicant pollution of the estuary, too. It is of a common recognition in the scientific society that suspended particles are scavengers for various dissolved toxicants (heavy metals and petro-hydrocarbons) in the coastal waters, particularly the fine particles with diameters less than micrometers being the major media for adsorption of heavy metals and absorption of organic toxicants due to their large specific surface areas. In the estuary region, there usually exist high saline gradients and high energy dissipation which lead to coagulation and sedimentation of fluvial fine particles there so that the heavy metals and organic toxicants attached to the fine particles go to the sediment together with the coagulated particles. So the sediment in an estuary is usually a sink for these toxicants where the toxicant compounds are accumulated and immobilized.

With rapid economic development, the coastal waters of South China Sea faces increasingly environmental stresses, one of which is organic and inorganic toxicant pollution in water and sediment phases, specifically involving persistent organic pollution and heavy metals. The persistent organic pollution can be subdivided into polycyclic aromatic hydrocarbons PAHs, polychlorinated biphenyls PCBs, and organochlorine pesticides such as DDTs and HCHs. The field monitoring and assessment of these toxicant pollutants have been widely made since 90s, particularly in Macau coastal waters^{[1][2]}, Victoria Bay of Hong Kong^[3], Xiamen Bay^{[3][4]}, as well as the whole Pearl River estuary^{[5][6][7]}. These studies were focused on field sites sampling of water and sediment and biota samples, and laboratory atomic absorption and chromatographic analysis of the samples for quantifying contents of the toxicant species in water and sediments. One of the research objectives of these studies was to trace back to the pollutant origins by collecting and analyzing the field monitoring data, which may includes atmospheric input, long distance fluvial transport, as well as local waste dumping (municipal wastewater, urban runoff).

The Pearl River delta is situated in the lower reaches of the Pearl River network which includes West River, Pearl River and East River as well as lots of tributaries connecting these main rivers as a net. Macau is just located at the west tip of the delta. The Macau coastal waters looks like a giant sink of suspended solids converging most of the solids transport fluxes through the west delta water network and in the same time collecting large quantities of organic and inorganic toxicants attached to the suspended particulates and accumulated in the coastal water sediments annually. It has been already recognized that most of the persistent organic pollutants in water environment are hydrophobic and preferring to staying in particulate phase instead of water phase. High affinity of the heavy metals pollutants to particulate phase has also been widely recognized. Therefore the sediments of the coastal waters become a sink of the organic and inorganic toxic pollutants from the upstream regions as well as the local sources. The Macau coastal waters are characterized by high turbidity with high suspended solids levels and active pollution interactions at water/sediment boundary. Therefore, the physicochemical interactions and exchange processes between water and particulate phases are the key for understanding fate and pathways of the toxic pollution of the coastal waters, which involve heavy metals accumulation and possible remobilization of the sediments^{[8][9][10]} and persistent organic pollutants transport and transformation in the sediments^{[11][12]}.

1. Field Monitoring Assessment of Toxic Pollution in Sediments^{[1][2][13]}

In March 1997, the first field monitoring program was made over the Pearl River estuary and its upstream waterways. Totally 17 sampling sites were selected to collect surface sediment samples with depths of 10-20 cm, including Guangzhou harbor and Macau coasts. The persistent organic compounds PAHs, PCBs and organochlorine pesticide residues in the samples were measured in

laboratory by GC-MSD and GC-ECD chromatographic analytic procedures. It was found that one of the highest persistent organic concentrations occurred in the surface sediments sampled at Porto Interior of Macau (ZB13) which is a sink of suspended fine particles transporting from the upstream waterways. Because of affinity of the hydrophobic organic compounds PAHs and PCBs to solid phase, the fine particles deposition led to accumulation of the toxic compounds in the sediments of Macau. The atmospheric dry deposition might be of another source of the organic pollutants accumulation in the sediments. Table 1 presents the details of the field monitoring results.

Table 1 Concentrations of organic contaminants in sediment samples ^[1]

Sample number	ZB07	ZB08	ZB09	ZB10	ZB11	ZB12	ZB13
Sampling sites	Lingdingyang Bay						Macau Porto Interio
	Wanqingsha	Shenzhen bay	Lingding island	Qiao	Jiuzhou Bay	Macau Nam Van Lake	
Organochlorinated pesticides							
HCHs	1.6	0.14	1.45	2.64	1.65	2.36	2.85
DDTs	10.17	2.6	12.31	10	10.63	115.61	1628.81
Other pesticides	5.98	2.89	3.87	9.68	8.09	11.46	26.52
Total pesticides	17.75	5.63	17.63	22.32	20.37	129.43	1658.18
Polycyclic Aromated Hydrocarbons PAHs							
Naphthalene (Na)	55.77	10.77	32.81	40.22	43.72	43.29	50.15
acenaphthene	10.84	nd	nd	nd	43.72	43.29	3.58
acenaphthylene	nd	nd	nd	nd	nd	nd	7.16
Fluorine (Fl)	54.22	nd	15.44	65.36	87.43	86.58	32.24
Phenanthrene (Ph)	109.3	11.24	60.15	65.36	65.57	64.94	136.69
Anthracene (An)	10.41	nd	5.47	5.45	5.46	5.41	20.63
Fluoranthracene (Flu)	33.83	5.5	20.41	21.28	25.66	25.41	247.58
Pyrene (Py)	26.02	5.5	23.33	18.24	14.26	14.12	221.79
Benzo(a)anthracene	23.42	5.5	14.58	9.12	8.55	8.47	162.48
Chrysene (Chr)	62.46	11	35	39.52	37.06	36.7	232.11
Benzo(b)fluoranthracen	54.65	22.48	28.87	130.72	206.09	204.08	1786.87
Benzo(k)fluoranthracen	31.23	22.48	18.37	65.36	131.15	129.87	1326.33
Benzo(a)pyrene (BaP)	31.23	11.24	18.37	43.57	74.94	74.21	1492.12
Indene(1,2,3-cd)pyrene	33.83	22.48	21	87.15	112.41	111.32	1326.33
Dibenzo(a,h)anthracene	26.02	5.62	10.5	43.57	56.21	55.66	957.91
Benzo(ghi)pyrene	36.43	22.48	26.25	98.04	93.68	92.76	1215.81
16 TCL PAHs	599.68	156.32	330.55	732.96	1005.91	996.11	9219.78
2-3 ring PAHs (%)	40.11	14.08	34.45	24.07	24.45	24.45	2.72
Total PAHs*	2284.1	323.07	995.76	1959.96	2372.16	2349.06	14811.5

*: Total PAHs include parent PAHs, Alkyl-PAHs, N/S – PAHs (μ g/kg, dry weight)

Therefore, the Macau coastal waters has become the highly ecologically risky water zone of the persistent organic pollution because of high accumulation of these compounds in the sediments of the coastal waters. In order to further clarify the highly risky persistent organic pollution in the coastal waters, particularly to find out specific sources and pathways of specific persistent organic compounds, the follow-up field monitoring program was made in October 1998 with 45 sampling sites located around the coastal waters (e.g., at Porto Interior, Porto Externio, Nam Van Lake, Coloane water area, etc.) and the surrounding waterways such as Qianshan river, Shizimen waterway, Maliuzhou waterway, Jiuzhou port. The persistent organic compounds in the surface sediment samples were analyzed and measured by the same procedures in laboratory as it did in March 1997. It was found that the distribution profiles of unsubstituted PAHs or parent PAHs (P-PAHs) among these sampling sites were following the similar patterns. The P-PAHs analyzed included benzo[e]pyrene(BeP), benzo[a]pyrene(BaP), benzo[b]fluoranthene(BbFlu), inde[1,2,3-cd] pyrene (In[1,2,3-cd]P). The

monitoring data clearly indicated that these PAHs pollutants came from the identical source through the similar pathways to the coastal waters, i.e., fluvial transport of suspended solids with highly accumulated PAHs. It is some sort of non-point pollution source initiated over large areas of the upstream delta.

The PAHs contents in the surface sediment samples at Porto Interior sites (MC25-34) were found to be in the range of 4000-7000 ng/g dry weight dramatically higher than these of the other sites (1000-2000 ng/g dw), which indicates that there exists another local pollution source leading to the extremely high PAHs pollution level in the Porto Interior samples. It is believed the source was of Macau urban runoff and municipal wastewater discharge into the water zone surrounding Porto Interior. Table 2 presents the details of the field monitoring output.

Among these samples taken at sampling sites except Porto Interior, the values reflecting the dependencies of the ratio of Alkyl-PAHs/P-PAHs vs. the ratio of low molecular weight (LMW)/high molecular weight (HMW) were found to be close each other (1.3-2 vs. 0.6-1.2) while the dependencies of these two ratios at Porto Interior showed a different picture with quite scattering of the ratio dependency values, which further confirmed existence of the local pollution source near Porto Interior. Another dependency between the ratios of BaP/BeP and BaA/Chr were studied among these sample sites, too. The BaP/BeP and BaA/Chr ratio dependency reflects bio-degradation of the PAHs compounds in water environment, which can also be used to indirectly identify the pollution sources and pathways. The ratios of BaP/BeP for the Porto Interior sediment samples were found to be close to these of the other sites, which means that the high rings PAHs (>5 rings) pollution sources are identical among these sites including Porto Interior. On the other hand, the ratios of BaP/Chr at Porto Interior were much higher than the other sites, which means that for the low ring PAHs (< 4 rings) pollution, Porto Interior is near the local point pollution source and the other sites are far away from the source.

By further analyzing the chemical compositions of the low and high ring PAHs in the samples from the different sites and on comparison with the PAHs compositions of aerosol and dust in ambient air samples of Macau, the third pollution source of the PAHs in the coastal waters was identified by this field monitoring study, i.e., atmospheric dry deposition. Figure 1 a, b, c, d present the 3-4 rings P-PAHs and 5-6 rings P-PAHs compositions, respectively among the samples of Porto Interior and the other sites in the coastal water sediments as well as the aerosol and dust samples of ambient air of Macau. It was found that the low ring (3-4 rings) PAHs compositions were quite different among these samples while the high ring (5-6 rings) PAHs compositions are similar between the air particulate and water sediment samples including these of Porto Interior, which means that part of the high ring PAHs came

Table 2 Field Monitoring Results for PAHs in Sediments (ng/g, dw)^[2]

	Na	Fl	An	Ph	Py	Flu	BaA	BaP	P-PAH	A-PAH	O/S-PAH
qMC43	12.6	10.2	3.1	48.1	27.8	29.2	7.3	14.1	224.1	342.6	43.4
qMC42	22.7	34.1	15.5	164	81.8	86.2	39.4	40.9	800.9	1265.8	161.4
qMC41	0	8.5	4.7	43.4	35.1	33.6	14.6	11.3	232.4	373.6	63.7
qMC40	0	8.5	7.8	72.9	64.3	59.9	39.4	29.6	534.1	794.7	134.2
qMC39	2.5	25.6	7.8	96.1	57	55.5	29.2	28.2	532	983	163.7
qMC38	37.9	23.9	10.9	89.9	67.2	59.9	27.8	22.6	541.9	928.7	169
qMC37	42.9	30.7	14	110	74.5	78.9	40.9	31	707.2	1159.5	194.4
qMC36	15.2	35.8	23.3	178	115	129	73	49.3	1025.6	1462.8	260.9
qMC35	40.4	25.6	12.4	89.9	70.1	61.4	46.8	33.8	692.7	1364.9	249.6
qMC34	27.8	23.9	14	79.1	57	46.8	27.8	28.2	551.2	1104.2	175.8
iMC25	32.8	29	31	135	208	226	127	94.5	1676	1794.8	371.9
iMC26	53	46	27.9	167	189	118	71.6	40.9	1043.3	3558.8	792.3
iMC27	20.2	17	27.9	126	260	301	234	161	2658.9	1022.1	216.2
iMC28	42.9	54.5	34.1	178	304	254	158	97.3	1940.2	3569.6	697.2
iMC29	48	40.9	23.3	126	142	126	107	66.3	1247.6	2303.6	433.1
iMC31	50.5	46	15.5	133	127	110	62.8	40.9	1012.7	2700.9	575.2

iMC30	50.5	44.3	12.4	150	136	110	61.4	45.1	962.4	2691.8	650.1
iMC33	53	186	94.6	1002	763	722	218	111	4178.4	2692.3	498.1
iMC32	17.7	30.7	15.5	99.2	87.7	80.4	55.5	47.9	836.2	1346.6	247.2
sMC11	27.8	27.3	18.6	158	118	108	39.4	33.8	874.7	1251.3	220.2
sMC12	40.4	18.7	9.3	101	52.6	48.2	24.8	22.6	513.2	860.1	140
sMC13	58.1	18.7	9.3	82.2	36.5	35.1	17.5	18.3	432.9	820.1	106.2
mMC45	30.3	18.7	17.1	126	104	95	32.1	24	654.2	842.7	176.8
mMC47	50.3	25.6	9.3	123	71.6	70.1	24.8	22.6	600.6	963	165.8
mMC24	12.6	3.4	1.6	15.5	7.3	5.8	4.4	2.8	91.8	153.6	15.1
mMC23	15.2	5.1	3.1	20.2	14.6	16.1	10.2	8.5	175.6	170.2	22.5
nMC14	30.3	22.1	10.1	87.6	40.2	37.3	22.6	28.2	470.8	1012.3	122.6
nMC15	48	11.9	4.7	57.4	20.5	20.5	10.2	9.9	283.3	492.9	65.3
nMC16	42.9	32.4	17.1	152	95	87.7	29.2	24	738.2	1247.7	219
nMC17	17.7	23.9	12.4	119	101	81.8	33.6	29.6	680.9	1307.3	254.4
nMC18	15.2	8.5	4.7	40.3	24.8	23.4	11.7	9.9	232.9	347.8	60.8
nMC19	17.7	23.9	9.3	119	57	49.7	14.6	12.7	461.6	1050	147
nMC22	0	13.6	10.9	107	68.7	61.4	20.5	18.3	499.4	871.1	178.8
nMC21	25.3	23.9	14	119	70.1	64.3	24.8	19.7	569	1147	202.1
nMC20	15.2	25.6	14	144	99.3	90.6	27.8	25.4	682.8	1108.9	214.5
cMC08	15.2	10.2	4.7	45	24.8	24.8	7.3	5.6	220.8	353.5	48.3
cMC09	22.7	22.1	17.1	127	87.7	80.4	27.8	26.8	680.7	975.9	171.5
cMC10	73.2	22.1	12.4	115	67.2	65.7	35.1	33.8	682.1	922.8	150.4
jMC01	40.4	17	9.3	79.1	40.9	36.5	24.8	29.6	475.9	784	113.9
jMC02	7.6	17	7.8	82.2	40.9	35.1	26.3	22.6	457.5	801.9	122.8
jMC03	45.5	15.3	7.8	69.8	36.5	30.7	23.4	18.3	450	859.8	116.8
eMC06	53	25.6	9.3	99.2	48.2	43.8	32.1	24	567.6	1050.4	136.4
eMC04 ^a	30.3	15.3	7.8	72.9	35.1	32.1	23.4	14.1	413.5	649	87.4
eMC05	104	30.7	18.6	163	92	87.7	45.3	35.2	886.8	1237.8	191.7
eMC07	50.5	20.4	9.3	85.3	43.8	39.4	23.4	18.3	460.8	846.3	118.4

Note: qMCxx – Qianshan River; iMCxx – Porto Interior; sMCxx – Shizimen Waterway; mMCxx – Maliuzhou Waterway; nMCxx – NamVan Waterway; cMCxx – Coloane Waters; jMCxx – Jiuzhou Port; eMCxx – Porto Exterior. Na – naphthalene, Fl – fluorine, Ph – phenanthrene, An – anthracene, Flu – fluoranthrene, Py – pyrene, BaA – benzo(a)anthracene, BaP – benzo(a)pyrene, P-PAHs – parent PAHs, A-PAHs – alkyl-PAHs, O/S-PAHs – oxygen/sulfur PAHs, T-PAHs – total PAHs

from the atmospheric dry deposition and the PAHs transfer at water/air boundary is one of the important pathways of the PAHs pollution in the coastal water sediments, particularly for the high ring PAHs.

By further analyzing the PAHs compositions in the sediment samples, it was found that the PAHs pollution was constituted by three main PAHs groups caused by three different pollution sources, i.e., group I Alkyl-PAHs and O/S PAHs counting for about 30% of the total, which reflects petroleum pollution related to the urban runoff from the Macau city; and group II high molecular weight PAHs (228-302) counting for 27% and resulting from high temperature combustion of fuels. The composition was close to that of the aerosols in ambient air. It may come from vehicular exhaust emission of the Macau city through the atmospheric dry deposition. The group III was of low molecular weight PAHs counting for 18% of the total which are products of low temperature combustion of oil and may be input by fluvial transport from the upstream delta. The remaining PAHs included 6.5% volatile polycyclic naphthalene and dimethylnaphthylene and 2% PAHs species from

geological mineralization in nature.

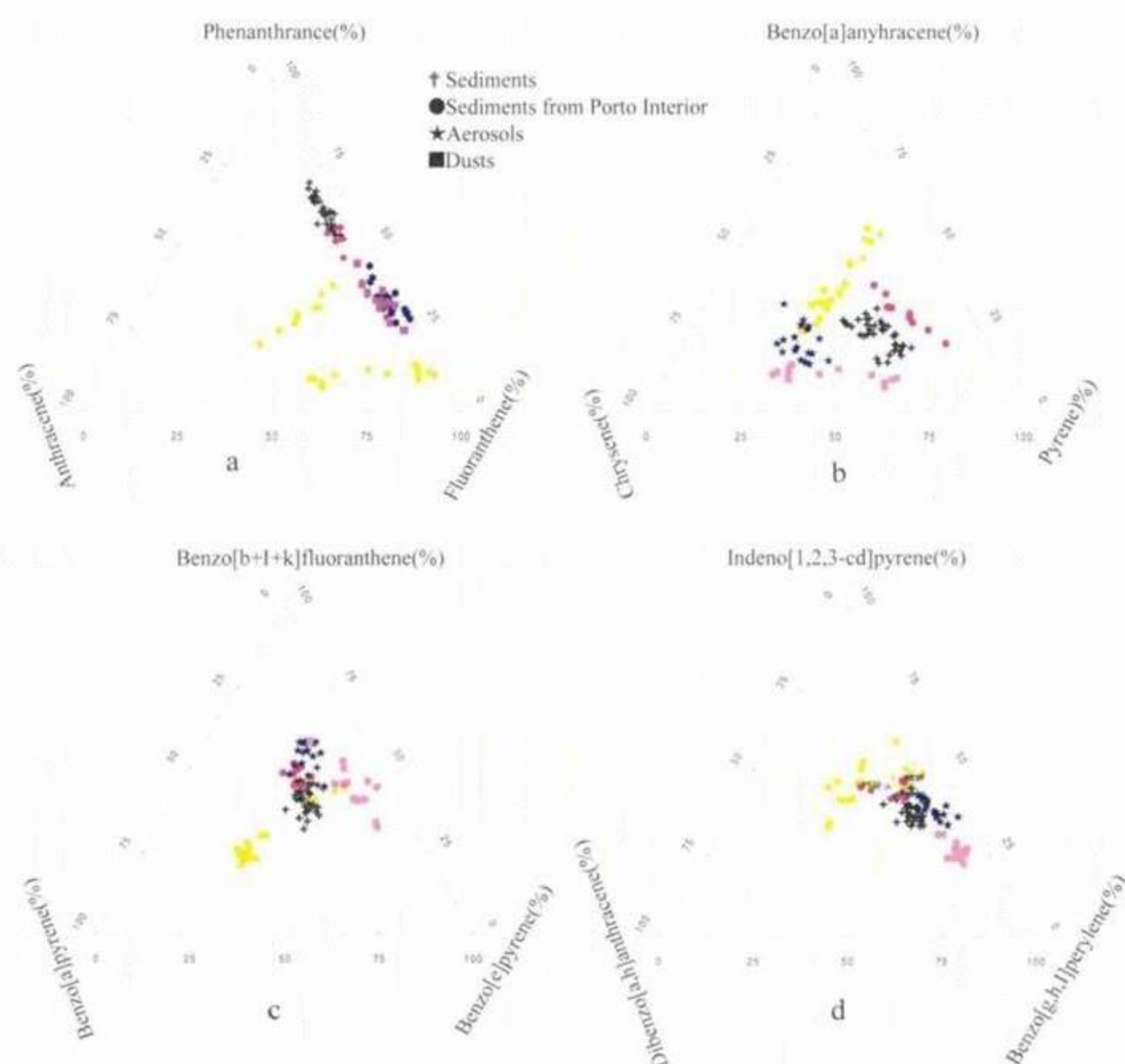


Figure 1 P-PAHs chemical compositions in sediments and aerosols in Macau ^[13]

The laboratory affiliated to Macau Medication Bureau has made a long term field monitoring program since 1992 for monitoring the annual variation of heavy metals in the surface sediments of the Macau coastal waters. Table 3 gives the 1997 monitoring results ^[9] of the heavy metals Pb, Cd, Cr and non-metal As in the sediments. It was found that the partitioning coefficients (K_d) were 7,600 - 20,000 l/kg among these species at interfaces between pore water and sediment, which means that the pollution species have been already accumulated in the sediment as the persistent organic pollutants (e.g., PAHs) were.

Table 3. Monitoring results of heavy metals and other toxic species in the water phase (W) and the sediment phase (S) ^[9]

	water	solid	water	solid	water	solid	water	Solid
Porto Interior 1		8.4		44		4		26
Porto Interior 2	0.04	1.8	2	35	1.1	36	2.3	22
Areia Preta	0.2	1.8	3.5	28	3.5	37	2.15	20
Praia Grande		4	2.5	45	1.3	29	3.6	22.5
Coloane	0.1	0.6	6.5	15	2.8	24	3.25	17.5
Pac On		6.2		37		35		31.6
K_d (l/g)	20		11.3		18.8		7.6	

In March 1997, a sediment column with 70 cm depth was taken at Nam Van Lake for measuring variations of heavy metals contents along the column depth. The heavy metal contents then were measured in laboratory by atomic absorption procedure. The heavy metals involved were Pb, Cu, Zn, Ni, Cr and Cd as well as As. By the ²¹⁰Pb measurement, the sediment column was aged to estimate annual variation of the heavy metal species in the period of 60s to 90s. It was found that there were

peak contents of these heavy metal pollutants occurring in 90s, which is just a period of rapid economic development of the Pearl River delta. It has been recognized that the estuary sediment is not only a sink for heavy metal pollution but also a second source for heavy metals 'fixed' in the sediment releasing back to the water body. The sediment 'fixed' heavy metals may be remobilized from the sediment if the environmental chemical condition at water/sediment interfaces changes due to coincidence of other pollution events. For example, overloading of degradable organic and nutrient wastes from sewage at the interface may lead to anoxic environment there and anaerobic acidification of organic wastes will produce various volatile fatty acids accumulated in the surface sediment [14]. Some physical processes occurring at the interface can also cause metal remobilization. For example, resuspension of the surface sediment resulting from high shear stress near shores may dramatically change the pH and redox conditions, leading to metal release from the sediment. It has been shown that the pH change and periodic redox cycles can shift the metal binding fractions on the sediment particulate surfaces to more mobilizing and soluble ^[15].

2. Physicochemical Modeling for Toxic Pollution in Water and Sediment Phases

Undoubtedly the field monitoring data can directly reflect the real world but a complex world. The toxic pollution in the coastal water sediments is a quite complicated problem involving various different pollution sources and pathways such as air/water exchange, surface adsorption and absorption, suspended particulates settling and transfer together with toxicants attached, resuspension of surface sediments, as well as long distance suspended solids transport in the tide flow field of the coastal waters. Only depending upon the field monitoring study, one cannot clearly understand the pollution fate and pathways. One of the reasons is that contaminated waters and sediments contain a mixture of aliphatic and aromatic organic compounds, each differing in its reactivity, solubility, toxicity, volatility, mineral surface affinity, and biodegradability. Another reason is that the suspended particulate matter and the sediment in natural systems are of a mixture containing phytoplankton, detritus, and clays and silts with different organic carbon contents and wide ranges of particle sizes. These different adsorbents do not behave independently. The accessibility of certain sorption reservoirs is probably influenced by the presence of others, and depends on the structure of the particles. The third reason may be that the hydrophobic organic compounds partitioning between suspended and solution phases is not always at equilibrium in water column. The field monitoring data are just reflecting the apparent consequence of interactions among these complicated processes. To clarify the fate and pathways of so many toxicant pollutions (various persistent organic species and heavy metals), physicochemical modeling for these processes and pollutants is essentially needed so as to imitate the reality by stressing those aspects that are assumed to be important and omitting all properties considered to be nonessential.

Bioavailability and ecotoxicology of the persistent toxicants (inorganic and organic) are closely related to their physicochemical transformation and partitioning between the overlying water phase and the suspended solid phase as well as the surface sediment phase. At present the laboratory assessment studies in the process mechanisms are still the most important tools to assess the impacts of these trace species on aquatic ecosystems of the coastal waters. The fundamental understanding of the process mechanisms is based on the chemical equilibrium concept, i.e., the equilibrium models for partitioning between the water and the sediment phases. In order to verify the validation of the laboratory-established models, the field monitoring for typical pollutants and sediments is necessary. Sometimes, considerations of the process kinetics are indispensable to extrapolate the laboratory study output to the actual cases.

2.1. Modeling for Water-Solid Phase Partition of Organic Compounds

Examining field monitoring data in Table 2 for various P-PAHs contents in surface sediment samples at different sites, one can see that the contents are only related to specific compounds in spite of different sites with different sediment particulates. The concentrations of different PAHs compounds in the sediment particulates vary in certain manner from the PAHs with low rings and low molecular weights to the high rings and high molecular weights. In order to insight the phase partitioning and distribution between pore water and sediment particulates, the partitioning coefficient

K_d is defined for a single compound (e.g., pyrene) to measure the compound phase distribution between pore water and sediment particles:

$$K_d = C_s/C_w \quad 1)$$

Where C_s - concentration of the compound partitioning to the particulate phase, ng/g; C_w - concentration of the compound in pore water, ng/l.

Chemically, the persistent organic pollutants in natural water environment are mostly of neutral and nonpolar organic compounds constructed primarily from carbon, hydrogen and halogen (e.g., Cl) atoms such as PAHs compounds. Most natural minerals such as sediment particulates are polar and expose a combination of hydroxyl- and oxy-moieties to their exterior^[16]. Naturally, then these polar surfaces strongly favor interactions which allow them to form hydrogen bonds with polar water molecules, and unfavorably directly adsorbing the nonpolar organic compound molecules such as pyrene and other PAHs on the natural mineral surfaces. In other words, replacing the water molecules at such a surface by nonpolar organic molecules is unfavorable from an energetic point of view. On the other hand, penetration of neutral organic chemicals into any natural organic matter such as humic substances in sediments does not require displacement of tightly bound water molecules. In light of this discussion, we may not be too surprised to find that nonreactive, neutral organic chemicals like PAHs show greater solid-water distribution ratios (K_d) for soils and sediments that contain high amounts of natural organic matter. The neutral and nonpolar organic compounds are absorbed in the natural organic matter attached with sediment particulates instead of directly adsorbing to the mineral surfaces of the sediment particles. Therefore, another partitioning coefficient K_{om} is defined to measure allocation of the neutral organic chemicals between pore water and the natural organic matter in the sediment:

$$K_{om} = C_{om}/C_w \quad 2)$$

Where C_{om} - concentrations of the compound absorbing in the natural organic matter e.g., humic substances in the sediment particulates, ng/g. The relationship between K_d and K_{om} is given by

$$K_d = f_{om}K_{om} \quad 3)$$

Where f_{om} - fraction of the natural organic matter content in the sediment particulates, %. The organic matter-water partition coefficient K_{om} is solely of an organic matter's property with no difference to different sediment particles. It has been found that the K_{om} values for a single neutral nonpolar chemical sorbing to a variety of soils and sediments are almost the same [16]. The K_d values are solely related to the natural organic matter contents f_{om} ; the higher the f_{om} value, the larger the K_d value will be. The C_s values should follow the same pattern if the C_w values do not change too much.

Table 4 presents the analytic results for mineral compositions of the sediment particulates at different sampling sites of the coastal waters. We can see that the organic carbon fractions f_{oc} are almost the same for different sites ranging from 1 - 1.27 %, and the organic matter fractions f_{om} can be calculated by doubling the f_{oc} values. As a result, the K_d values for a single neutral PAH chemical e.g. naphthalene (Na) or phenanthrene (Ph) should be almost the same among these different sites. Referring to Table 2, one can see that the C_s values for a single PAH chemical are in the same level, indeed, no matter where it was sampled. For example, the monitored Na contents (C_s) among the different sampling sites are mostly in the limited range of 30-50 ng/g dw and for the Ph contents, the C_s values vary in limited range from 110 to 160 ng/g dw.

In a fashion analogous to sediment-water partitioning, we can envision the molecules of a given compound partitioning between the organic phase and the aqueous phase. This partitioning process is determined by the relative fugacity of the compound in each phase and at equilibrium may be described by a dimensionless equilibrium constant.

The equilibrium constant can be also named as n-octanol-water partition coefficient:

$$K_{ow} = C_o/C_w \quad 4)$$

Table 4 Analytical Result for Mineral Composition of Sediments

	$f_{oc}(\%)$	$f_{om}(\%)$	Silica(%)	Clay(%)	$\rho_s(\text{kg/l})$	ϕ	r_{sw}	$r_{swl}(\text{kg/l})$
qMC	1.27	2.54	40.4	48.2	2.2	0.7	0.94	
iMC	1.35	2.7	36.3	51.6	2.2	0.7	0.94	7.34×10^{-5}
sMC	1.09	2.2	44	49.5	2.2	0.7	0.94	
mMC	1	2	32	54	2.2	0.7	0.94	
nMC	1.01	2	42.9	46.9	2.2	0.7	0.94	
cMC	1.29	2.58	35.5	58.1	2.2	0.7	0.94	
jMC	1.15	2.3	26.7	67.3	2.2	0.7	0.94	
eMC	1.25	2.5	32	61.5	2.2	0.7	0.94	

Where C_o - the concentration of the compound in the organic phase, ng/l. Partially because of the choices of early workers [16] and the special amorphous nature of C4 to C10 alcohols, but mostly because n-octonal is a reasonable surrogate for many kinds of natural and environmental organic solvents, the n-octonal-water partition constant for a single nonpolar organic chemical such as pyrene is used to generalize the compound partitioning between any organic phase and the aqueous phase. The n-octonal-water partitioning coefficient has been investigated extensively. Because the attachment of a neutral nonpolar organic chemical on sediment particles is essentially of absorption process of the compound to the natural organic matter in the sediments, there is certain relation between the K_{om} and the K_{ow} for the single compound. An empirical equation has been established to quantify the relationship:

$$\log K_{om} = c \log K_{ow} + d \tag{5}$$

Where c and d are empirical parameters whose values depend upon different compound classes. Karichhoff [17] examined the relationship between K_{om} and K_{ow} for a series of the neutral nonpolar compounds (aromatic hydrocarbons, chlorinated hydrocarbons, chlirio-S-trazines, phenyl ureas) by compiling the relevant data from different sources, and found that generally the values of parameters c and d were almost invariable and $c = 0.82$ and $d = 0.14$ if the unit of K_{om} is l/kg, instead of l/g. Table 5 gives the values of the K_{ow} for a series of P-PAHs compound including the 2-ring PAH Na, and the 3-ring PAHs Fl, An, Ph, and the 4-ring PAHs Py, Flu, BaA as well as the 5-ring compound BaP. The calculated K_{om} values based on Equation 5) are also given by that table. One can see that the higher the molecular weight and ring numbers of the compound, the larger the K_{om} value it has, which means that the neutral nonpolar PAH compounds with high ring numbers and high molecular weights are more favorable to absorb into the natural organic matter attached to the sediment particulates than low ring and low molecular weight compounds. Combining Equations 3) and 5), one can get the following equation:

$$\log K_d = c \log K_{om} + d + \log f_{om} \tag{6}$$

Table 5 Calculated Partition Coefficients of P-PAHs

	M.W.	$-\log C_{sat}$	$\log K_{ow}$	$\log K_{om}$	$\log K_d$
Na	128.2	3.61	3.36	2.9	1.27
Fl	166.2	4.96	4.18	3.58	1.95
An	178.2	6.46	4.54	3.86	2.23
Ph	178.2	5.2	4.57	3.89	2.26
Py	202.3	6.17	5.13	4.35	2.72
Flu	202.3	5.93	5.22	4.42	2.79
BaA	228.3	7.31	5.91	4.99	3.36
BaP	252.3	8.22	6.5	5.47	3.84

MW – molecular weight of PAH compound; C_{sat} – solubility of the compound

With it, one can calculate K_d values for these P-PAHs compounds which are also given in Table 5. Armed with such a K_d parameter for a case of interest, we may evaluate what fraction of a compound is in water phase f_w in a volume containing both solids and water by

$$f_w = C_w V_w / (C_w V_w + C_s M_s) \quad (7)$$

Where V_w - volume of water in total volume V_t , l; M_s - solid mass in the same total volume, kg. So considering Equation 1), one have:

$$f_w = 1 / \{ 1 + (M_s / V_w) K_d \} = 1 / (1 + r K_d) \quad (8)$$

Where r - solid mass concentration in the total volume, kg/l. For the surface sediment bed, the solid concentration can be expressed by the sediment bed porosity Φ , which is defined by pore water volume/total volume. That is

$$r = \rho_s (1 - \Phi) / \Phi \quad (9)$$

Where ρ_s - density of the solid, kg/l. The mineral compositions of the sediment particulates in the sampling sites are similar referring to Table 4. There are two major minerals silica and clay in the sediment particulates. Generally the silica counts for about 40% while the clay for 55%. The density of the sediment can be calculated by the weighting summary of silica density and clay density, which is 2.2 kg/l.

The compacted sediment bed porosity is usually 0.2 to 0.4, depending upon the particle size distribution. The larger the particle sizes, the larger the bed porosity values are. However, the surface sediment bed is not completely compacted. And there is a strong tendency of resuspension of the surface sediment due to strong shear stresses at the water/sediment interface. These both lead to a high porosity of the surface sediment bed, i.e., 0.7 - 0.8. With Equation 9), one can calculate the r value of the surface sediments in the coastal waters, which equals 0.94 kg/l. Substituting the K_d values in Table 5 into Equation 8), the f_w values for the P-PAHs species in Table 5 can be calculated. The calculation results show an interesting fact that most of the PAHs are partitioning into the sediment phase while in pore water the PAHs are almost undetectable ($f_w = 0$) except the 2-ring PAH Na ($f_w = 0.05$). The higher the ring number and molecular weight, the favorable the compound is to partition into the sediment phase.

In April 2001, the water samples were taken at the water depths of 0.5, 1.2, 3.5, 4.8, 5.5 and 6 meter (bottom), respectively along a water column at Porto Interior, aiming at analyzing partitioning of the persistent organic compounds (organochlorine pesticides DDTs and HCHs, and 16 US EPA TCL PAHs) between the bulk water phase and the suspended solid phase in the column with different water depths. The suspended solid concentrations at different depths of the column were measured to be 1.7, 17.9, 42.5, 54, 58.5 and 73.4 mg/l at the depths of 0.5, 1.2, 3.5, 4.8, 5.5 and 6 meters, respectively. Based on the K_d and r values of different compounds and different depths, the f_w values were calculated by Equation 8). The calculation shows a fact just opposite to the case of water-sediment partition that most of the PAHs are partitioning to the water phase instead of the suspended solids, particularly for 2-ring P-PAH Na. The calculated f_w values for Na are all equal to 1 at different column depths, which means that the Na pollutants are in dissolved phase instead of particulate phase. On the other hand, the calculated f_w value at the bottom for the 5-ring P-PAH BaP is 0.66, that is, the aqueous BaP pollution only counts for 66% of the total pollution and there still remain 34% absorbing onto the suspended particulates. However, at the top of the column (0.5m depth) where the suspended solids was only 1.7 mg/l, the calculated BaP f_w value is 0.99, that is most of the BaP species are in dissolved phase at that depth.

The field monitoring data also show the same trend, i.e., all the 2-ring PAHs were in dissolved phase at these depths while most of the 5-6 ring P-PAHs in suspended particulate phase, particularly at the bottom of the column. The 3-4 ring P-PAHs were detected in both phases and the dissolved phase partition fractions decreased with the depth. The 4-ring PAHs more favored the solid phase than the 3-ring P-PAHs (please refer to Figure 2).

In light of the above discussion, we can conclude that for a certain neutral nonpolar compound like Na or BaP, the dissolved and adsorbed (particulate) concentrations per total volume of a water

column, C_d and C_p can be expressed as

$$C_d = f_w C_t \quad (10)$$

$$C_p = (1 - f_w) C_t \quad (11)$$

Where $C_t = C_d + C_p$, representing the total compound concentration in the water column. For low ring and low molecular weight compound at top layer of the column, the K_d value is small and the r value is also small, $f_w \rightarrow 1$ and $C_d \rightarrow C_t$ and $C_p \rightarrow 0$, i.e., the dissolved phase partition is dominant. On the other hand, for high ring and high molecular weight at the bottom of the column (large K_d and r), $f_w \rightarrow 0$ and $C_p \rightarrow 1$ and $C_d \rightarrow 0$, this means that the absorbing or particulate phase is dominant.

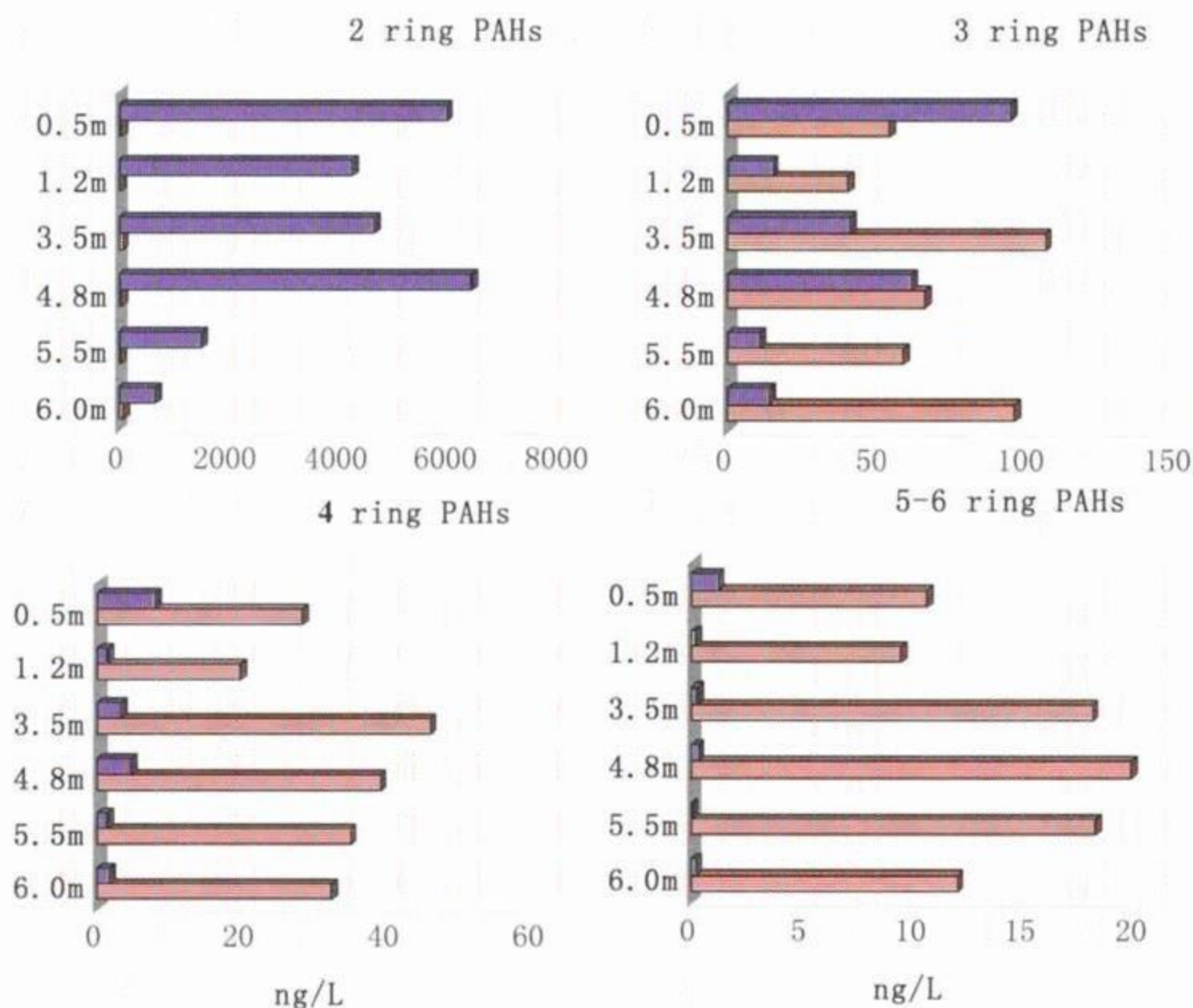


Figure 2. Field Monitoring Results for PAHs of Water-Suspended Solid Phases Partition in Water Column

(blue bar with vertical lines – dissolved phase; red bar with horizontal lines – suspended solid phase)

The conceptual model for the concentration of a persistent organic compound for which dissolved and particulate phases are both important can be established based on mass balance principle, i.e., the accumulation rate of the compound equals input rate subtracting output rate and interphase transfer loss rate as well as reaction decay rate. For the dissolved phase, we have

$$dC_d/dt = I_d - k_w C_d - k_g(C_w - C_a/K_h) - k_r C_d - J_{tr} \quad (12)$$

Where I_d - input rate of the compound in dissolved phase; k_w - output rate constant and $k_w = 1/t$ and t - water column detention time; $k_r C_d$ - reaction decay rate by hydrolysis, photolysis, biodegradation of the compound. Because most of the toxic organic pollutants are chemically persistent, this term can be omitted. J_{tr} - the compound transfer rate from the dissolved to the absorbing phases per unit volume. The term $k_g(C_w - C_a/K_h)$ represents the compound transfer from the water phase to the air, where K_h is the water-air partitioning coefficient of the compound and C_a is the compound mass concentration in the air. The air/water phase exchange rate constant k_g is determined by the compound molecular diffusion (D_w) and turbulent condition of the water/air boundary layers (e.g., wind speed at the water surface). For the sorbing or particulate phase, we have

$$dC_p/dt = I_p - k_w C_p - k_s C_p - k_r C_p + J_{tr} \quad (13)$$

where I_p - input rate of the compound in sorbing phase; $k_w C_p$ - output rate; $k_r C_p$ - reaction decay rate which can be omitted for most of the persistent organic pollutants. The term $k_s C_p$ represents the sorbing compound transfer rate with the particle settling to the bottom where k_s is the particle settling rate constant which is determined by particle Stoke's terminal velocity. Combining Equations 12) and 13), we have

$$dC_t/dt = I_t + k_g C_a/K_h - k_w C_t - f_w k_g C_t - (1 - f_w) k_s C_t \quad (14)$$

At steady state, i.e., $dC_t/dt = 0$, we have

$$C_t = (I_t + k_g C_a/K_h)/(k_w + k_g + (1 - f_w) k_s) \quad (15)$$

Where the air-water partitioning constant K_h is related to the Henry's constant by K_h'/RT and the Henry's constant K_h' representing air/water phase equilibrium. For most of the persistent organic pollutants such as PAHs, it is generally quite small (less than 10^{-3}). The air-water phase transfer rate constant k_g is equal to v_w/h , where v_w is the compound transfer velocity in the viscous layer at the water surface and related to the compound molecular diffusion constant D_w . For most of the persistent organic pollutants D_w is also small (less than 10^{-6} cm²/s). The water-solid transfer rate constant k_s is equal to v_s/h where v_s is the particle Stokes' terminal velocity. h is the water column depth.

2.2. Modeling for Heavy Metal Accumulation and Remobilization in Sediments

Table 3 gives the monitoring data for concentrations of Cd, Pb, Cr, and As species in the water phase and the sediment phase. A partitioning constant is defined to assess the accumulation of these heavy metals and the As species in the sediment as below:

$$K_d = [S \equiv M]/[M] \quad (16)$$

where $[S \equiv M]$ represents the concentration of the metal M in the sediment (μ g/g) while $[M]$ represents its concentration in water (μ g/l). The measured partitioning constants for these species were in 7.6 – 20 l/g. With Equation 8), one can calculate the f_w values for these species. For the water-sediment partitioning, the calculated values are all equal to 1, that is, all these pollutants accumulated in the sediment phase and undetectable in the pore water, just as the cases of the PAHs absorbing in sediments. For the water-suspended solid partitioning, the calculated values are in the narrow range near 0.5, i.e., the partitioning between the suspended and dissolved phases is just half to half.

The processes that govern the scavenging of trace metal elements by the particulate matter are complex, but generally can be considered to include adsorption, absorption, surface precipitation, and co-precipitation. In the oxic environment such as the oxygen-rich water column or the overlying suspended mud layers, the surface adsorption is dominant, and the allocation of the trace metals at water/solid interface can be estimated by the solid/water partitioning coefficients (K_d). The surface properties of the particles are an important key to understand interactions of trace metals and organic toxicants between dissolved and mineral phases in the coastal waters. The studies in particle surface characterization have suggested that an almost uniform coating of amorphous oxides or organic matter exists on the surfaces of the estuarine particles [18]. It is generally thought that iron and manganese oxides and organic matter coating are important phases for binding of trace metals in the sediment while clays can be only as a support for oxide and organic matter deposition or coating. The interactions of trace metals with the particle surfaces can be considered to be metal-ligand surface complex formations. The conceptual model can be established for interpreting the adsorption isotherms of trace metals at goethite oxide surface, based on the thermodynamic concept of surface coordination. The pH is a master parameter controlling the metal adsorption on oxide surfaces, and for a certain metal there is a special pH range where the adsorption dramatically increases.

Figures 3 gives experimental results of adsorption of Cu, Pb, Cd metal ions on goethite^[10], which show that there was a narrow special pH range for each of the metal ions that an adsorption jump occurred, and the adsorption increased gradually with the solution pHs while the desorption occurred when the solution pHs decreased.

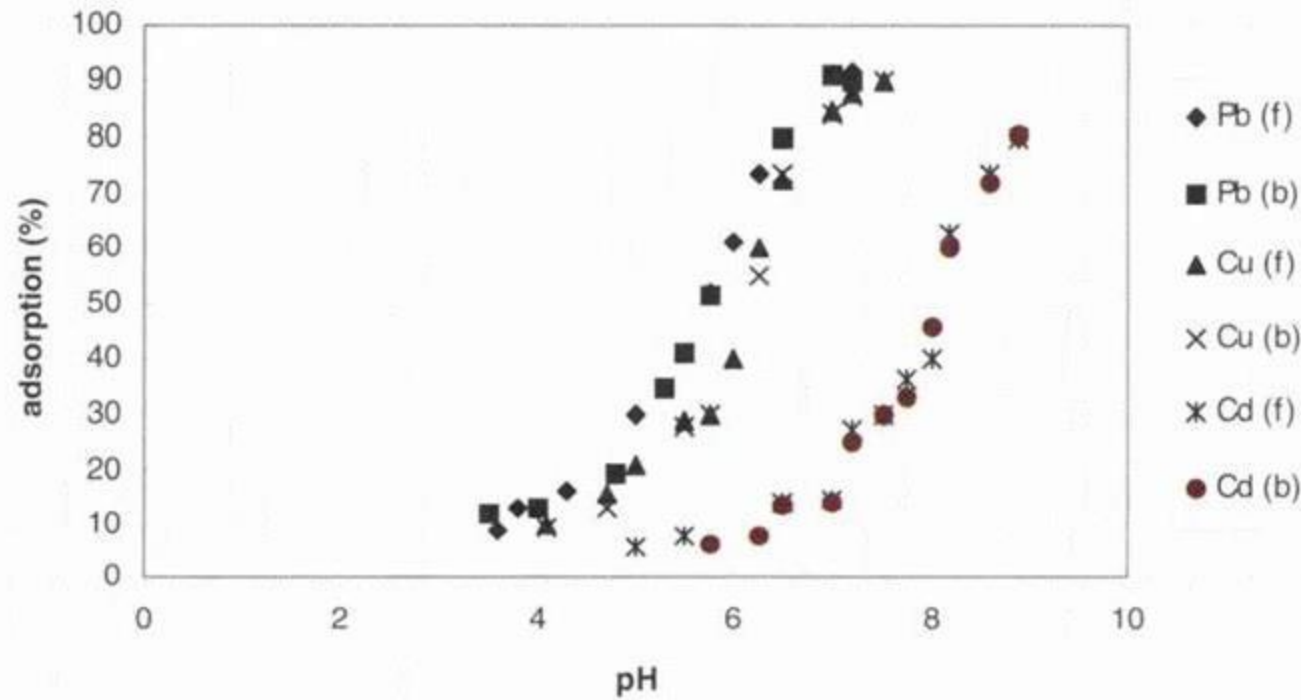


Figure 3. Adsorption (f) and desorption (b) of Pb, Cu, and Cd on goethite particles

This means that the adsorption is reversible with solution pHs. Viewed with chemical thermodynamics, and it can be considered as a process of metal-ligand surface complex formation, i.e.,



Where $S\equiv FeOH$ represents adsorption available site on goethite particle surfaces; $S\equiv FeOM^+$ is the particle surface site adsorbing metal ion M^{2+} .

At equilibrium,

$$K_s = ([S\equiv FeOM^+][H^+])/([S\equiv FeOH][M^{2+}]) \quad (18)$$

The log form of the above equation is given by

$$\log([S\equiv FeOM^+]/[M^{2+}]) = \log K_s + \log[S\equiv FeOH] + pH \quad (19)$$

Combining Equations 16) and 18), one can have

$$\log K_d = \log K_M + pH \quad (20)$$

which means that the partitioning between water and sediment is controlled by pH, and K_d is proportionally related to the pore water pH. Equation 20) shows such a linear relationship between $\log K_d$ and pH. Figure 4 was drawn based on the experimental data in Figure 3. The linear relationships for these metal adsorption of goethite were verified by the experimental data.

The interruption of the ecological cycling at the interface does not only illustrate a decrease of pH in the pore water but also an accumulation of dissolved organic matter which is mostly constituted by organic acids such as acetic acid. These organic acids have a strong affinity to complexing with the heavy metals such as Cu in solution, which can re-mobilize these metals already adsorbed on the particles. Figure 5 gives the desorption experiment for the Cu/goethite system.

This experimental result reveals a competition of the surface metal-ligand complexing ($S\equiv FeOM^+$) and the solution complexing (ML) for the water-copper ions-organic acids- goethite system, which can be expressed by the following equation:

$$\log\{[ML]/[S\equiv FeOM^+]\} = \log K - \log K_s + \log[HL^-] - \log[S\equiv FeOH] \quad (21)$$

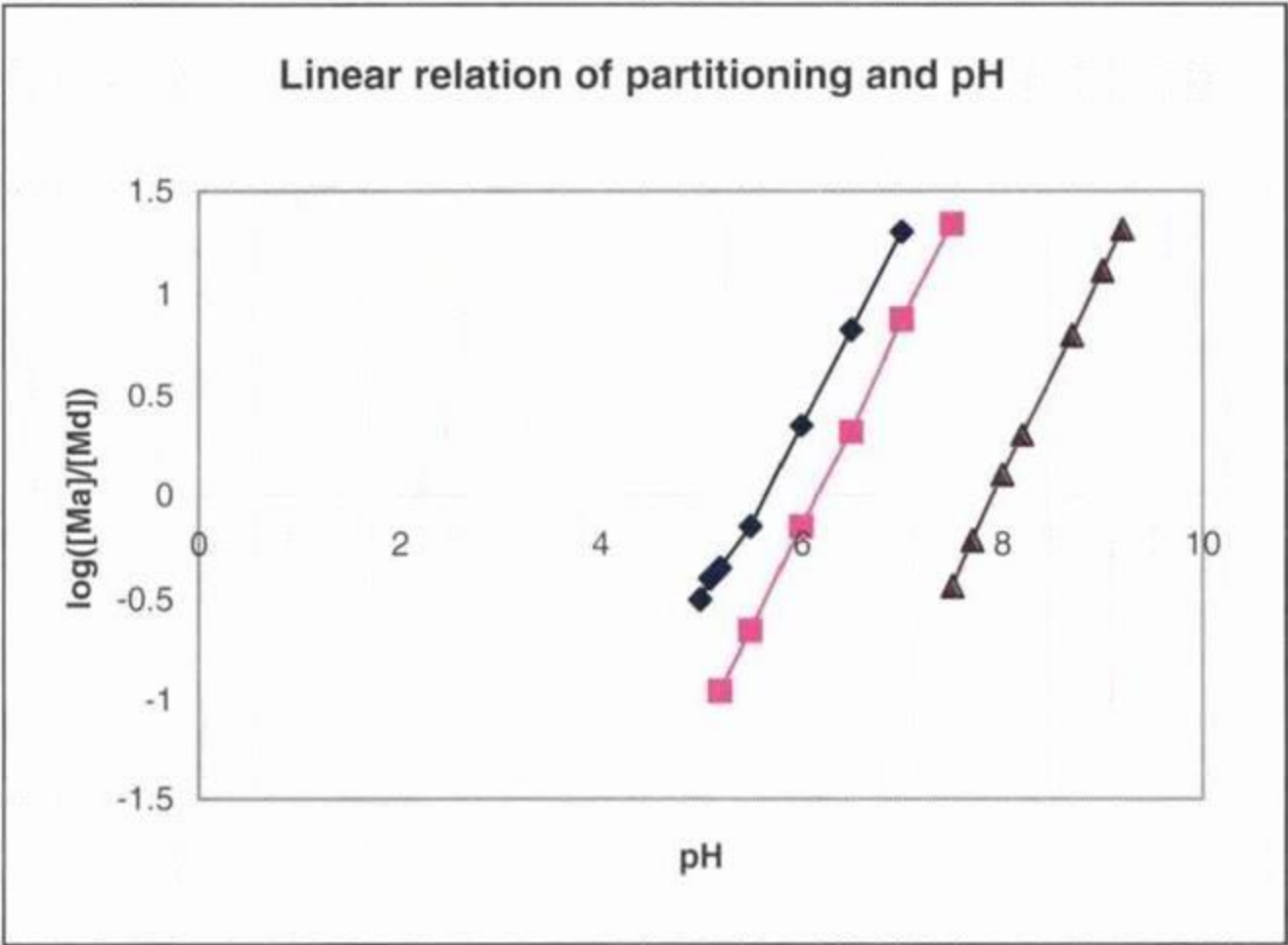


Figure 4 Linear relationship between partition coefficient vs. pH

which shows that the more affinity the organic acid with the metal ion such as citric acid, the more re-mobilizing the metal is. In Equation 21), HL^- represents the organic ligand, e.g., citric ligand and K is the solution complex formation equilibrium constant, and $\text{Log } K = 0.80, -1.14, -2.24, -1.51$ for citric, tartaric, salicylic and phthatic ligand complex formation with Cu(II) in solution, respectively.

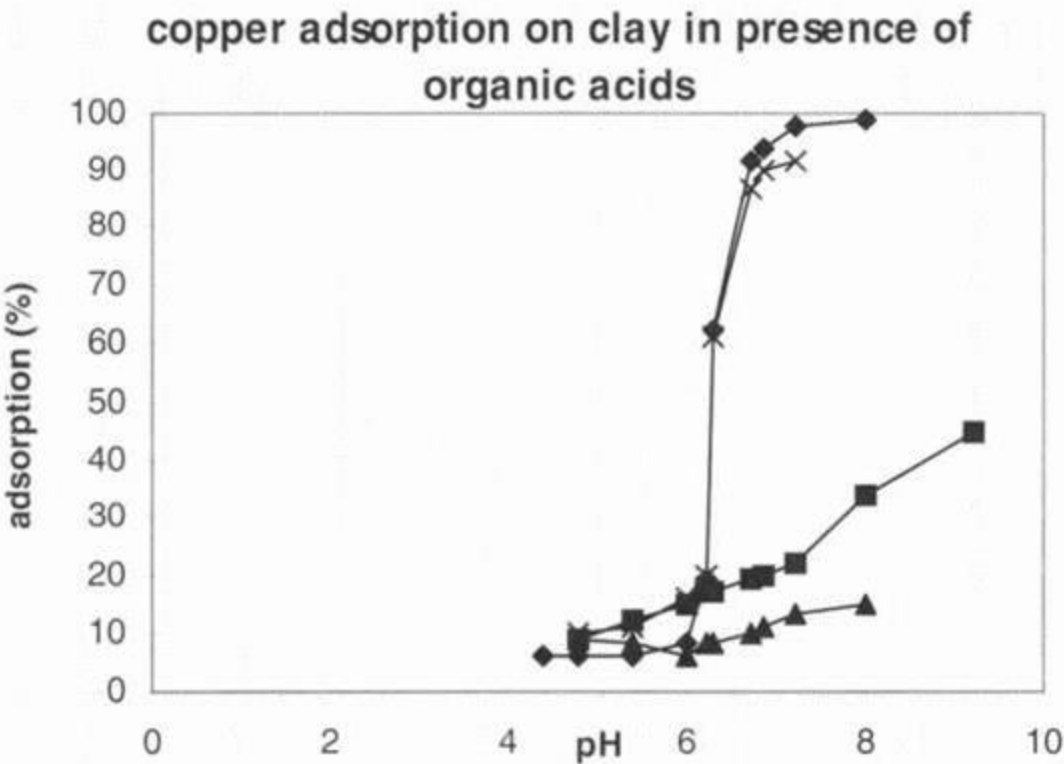


Figure 5 Copper adsorption on goethite particles in presence of citric(▲), tartaric – salicylic(■), phthatic(×) acids, respectively

Figure 6 gives the measurement of surface dissolution of goethite particles due to solution pH change and impact of organic acids (citric and tartaric acids). This shows that surface dissolution occurs at pH far from pH_{zpc} of goethite; and presence of organic ligands largely promotes such a dissolution process.

These phenomena can also be explained using the conceptual model of metal-ligand complex formation at water/particle interface. The mechanisms of proton- and ligand-promoted surface dissolution of goethite involve two steps: a fast step of the surface complex formation ($=\text{FeOH}_2^+$ or $=\text{FeL}^-$) and a subsequent slow step of release of ferric ions or ferric-ligand complexes from the goethite surface^[19]. The metal release can also be caused due to surface-controlled dissolution of goethite in presence of organic acids.

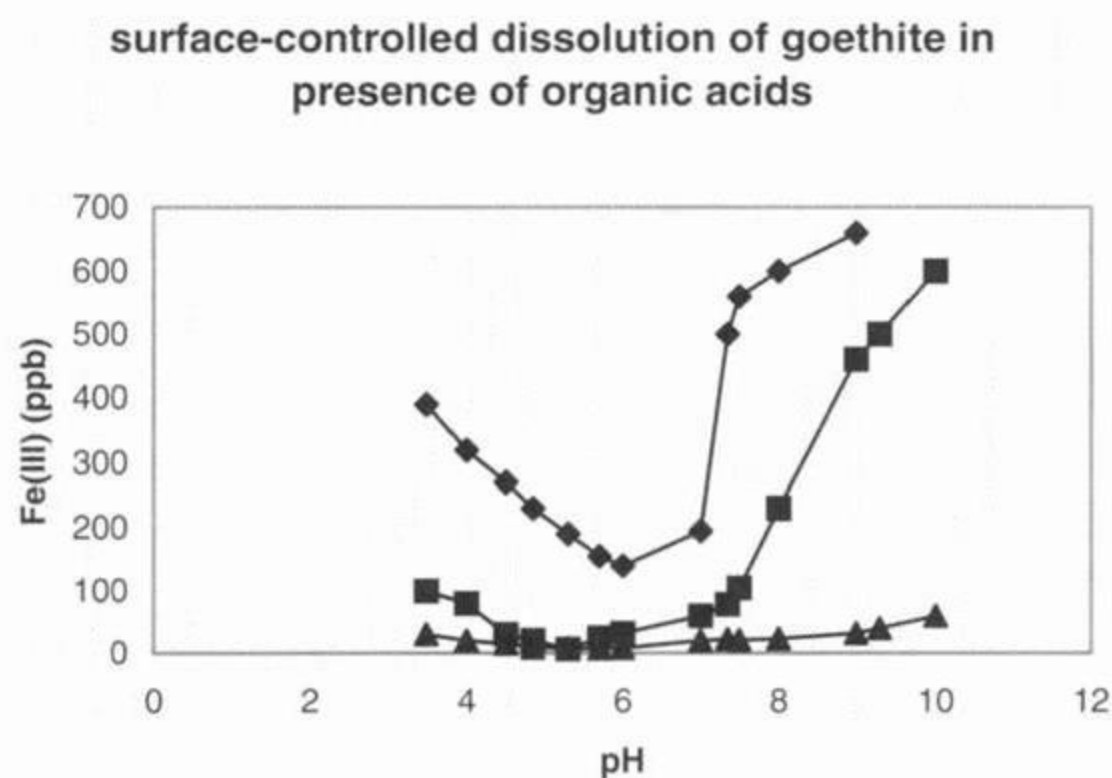


Figure 6 Surface-promoted dissolution of goethite in presence of organic acids

One of the mechanisms of metal re-mobilisation comes from the competitive effect between the metal-ligand complex formation in solution and the metal surface binding. The volatile fatty acids and humic acids that are the residues of organic matter degradation and decay as well as some synthetic organic compounds such as detergents, surfactants, tributyltin, etc. are all the available ligands for metals in the coastal waters leading to the metal re-mobilisation. Furthermore, some of the organic ligands such as acetic acids and salicylic acids can cause the surface dissolution of ferric oxide coated particles due to formation of solution complexes of these ligands with ferric ions in the lattice.

In this paper, the conceptual models are presented including the water-sediment partitioning model for persistent organic pollution in the Macau coastal waters and the surface complex formation model for heavy metal adsorption and desorption at water/sediment interfaces. All these models were established based on principles of organic and surface chemistry as well as the field observations. The model verification and calibration are further required by the designated laboratory experimental studies.

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Spatial Analysis of Vehicular Emission Pollution in Macau Peninsula

U Wa Tang, Zhishi Wang

(Faculty of Science and Technology, University of Macau)

Abstract: GIS (Geographical Information System) is a newly powerful tool for environmental assessment and analysis since the quality of environment is highly related to the “geographical” related factors. GIS can integrate such factors as “information” stored in a database within a “system” for better analysis. This article is focus on finding the spatial relationship between vehicle-dominated air quality and its geographical constrain, so called the street canyon effect in Macao peninsula. Spatial patterns of NO, NO₂, CO as well as canyon levels were modeled in front of each building block in main streets of Macao peninsula and apparent spatial relationships were found. Such findings verifies the significance of street canyon effect as an indicator for air quality assessment as well as a control parameter for urban planning which can contribute the decision making process for our government.

Keywords: urban spatial data, dispersion, air pollution, GIS

1. Introduction

Nowadays air quality of a city is reported according to the data collected from fixed monitoring stations at various functional areas such as the industrial area, residential area, road-side situation, as well as urban and suburban area etc. It may be suitable for simple geographic conditions where dispersion condition is simple. However, in urban area, such as Macao peninsula, with various microenvironments because of the difference of height of buildings and the complex traffic emission, one roadside monitoring station is insufficient to represent the whole function area.

A new approach using modeling technique to access air quality in front of each building block besides the main streets of Macao peninsula was targeted in this research. Evaluating suitable model under situations of Macao and then run all model cases automatically were the main stages. As traffic jam condition and street canyon dispersion condition are the two main characteristics controlling the air quality in Macao, two corresponding widely used mathematical dispersion models were then chosen for evaluation. Afterwards, it is clearly impossible to prepare all input spatial data files for each building block manually as there are more than 400 cases in Macao peninsula. GIS was then utilized successfully to overcome such time-consuming and human-error-inducing stage. Finally, the pattern of air quality in Macao peninsula was found varies according to its street canyon geometry.

2. Evaluation of Dispersion Models

There are many dispersion models with different complexity and different levels involving urban spatial data. Two models, CAL3QHC and OSPM, were evaluated. They represent different approaches of emission estimation and dispersion prediction for US and Europe urban situations. Comparison of them was given according to BERKOWICZ et al (1997), PAUL (1979) and US EPA (1995).

2.1. Comparison of Geographical Consideration

As CAL3QHC estimated the pollutant at intersection, geometry of all intersection lanes were involved. A series of receptors could be placed along the side of lanes. The height and location of the surrounding building blocks were not necessary and a user specified surface roughness coefficient was used. The range was from 0.03cm to 370cm representing smooth surface to residential apartment conditions. The dispersion estimation was based on the flat surface geographical environment.

OSPM focused on estimating a single urban canyon street. Only two receptors were used to represent the condition of windward side and leeward side in front of the study building block. The input parameters included the heights and locations of all nearby building block as well as the direction of the studying street. OSPM required more detail spatial data and hence, as shown later, proved to be more sensitive than CAL3QHC.

2.2. Comparison of Traffic Flow and Emission Estimation

CAL3QHC had a good improvement for estimation of the emission, which was separated into two parts at the intersection in signal junction by using the method in the Highway Capacity Manual in US. One was the emission from free flow condition during the green signal. The emission factor was user specified, which was modeled by Mobile 5 program of EPA according to the speed condition etc. Another was the queue condition during the red signal. By the approach traffic flow, the growing of the queue lane was estimated and the emission was calculated by the specified idle emission factor, which is much larger than the classical free flow emission factor. Estimation was hence carried on under the approach condition and the Under-Saturated/Over-Saturated Conditions.

OSPM did not require inputting the idle emission factor for the queue lane concept was not applicable here. However, the hourly average speed of the vehicle was needed and the corresponding emission variation for each type of vehicles was then adjusted.

2.3. Comparison of Dispersion Component

The dispersion component used in CAL3QHC was CALINE-3. CALINE-3 estimated air pollutant concentrations resulting from moving vehicles on a roadway based on the assumptions that pollutants emitted from motor vehicles traveling along a segment of roadway can be represented as a "line source" of emissions, and that pollutants were treated as dispersing in a Gaussian distribution from a defined "mixing zone" over the roadway being modeled.

OSPM used a simplified parameterization of flow and dispersion conditions in a street canyon that was deduced from extensive analysis of experimental data and model tests under different street configurations and a variety of meteorological conditions.

Concentrations of exhaust gases are calculated using a combination of a plume model for the direct contribution and a box model for the re-circulating part of the pollutants in the street.

That is, C_d and C_r are calculated separately as, $C_{st} = C_d + C_r$

Where C_{st} is the total concentration; C_d is the direct gauss plume dispersion by traffic emission; C_r is the recirculating part of the pollutants in the street.

2.4. Comparison of Model Results

Totally 3431 hours of monitor data from Aug. 1999 to Aug. 2000, excluding non-working days, were used to evaluate the dispersion model CAL3QHC and OSPM. Half-day monitor CO data from midnight to noon was shown in Fig.2. A clear pattern was shown that in 1-7 a.m. the minimum concentration was around 0.5 ppm and variation was less. After 7 a.m. the base line suddenly rose to 1 ppm and maintained in the following period with high variation. Such behavior was closely related to the traffic loading condition. Comparing to the CAL3QHC model values, a correlation of 0.46 was adopted. It was shown that the distribution of the modeled points lay lower than the monitor data in general. It is probably due to the model assumes flat topography as in highway typically found in US.

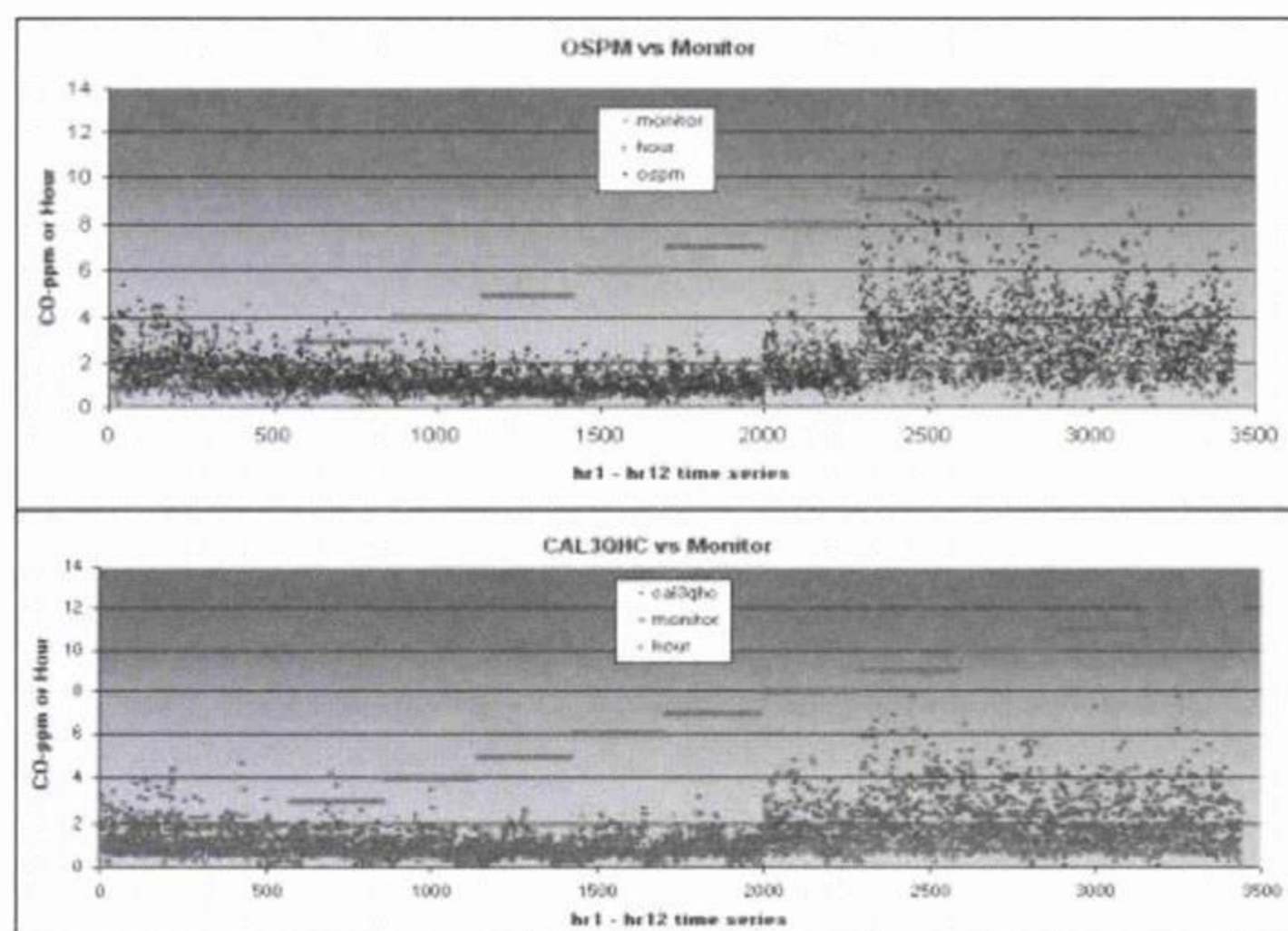


Figure 1 One year monitor vs. model pollutants by CAL3QHC and OSPM in hourly time series

For OSPM, although real time traffic-count data were not available and only substituted by a typical profile, relative good correlation was found between the modeled and the measured concentrations in both CO and NO_x. Fig.1 showed that OSPM behaved well in predicting the minimum and highest concentration in general. The correlation coefficient of model and monitor for CO and NO_x were 0.62 and 0.63.

Although CAL3QHC considered more in traffic jammed condition, the about results showed that utilization of the height of building and width of street spatial data with corresponding wind tunnel deduced dispersion parameters by OSPM was more sensitive.

2.5. GIS and OSPM Connection

The conceptual framework for this GIS/OSPM connection allowed programs within GIS to produce the necessary information for the input file of exterior model OSPM. The ArcView GIS software then executed user written internal program in order to obtain a properly spaced input file and executed the model. Finally, the model output was processed, imported back into GIS, and viewed as charts, tables, and coverages. Specifically for this research, the model input is read by the ArcView programming language Avenue from tables and coverages

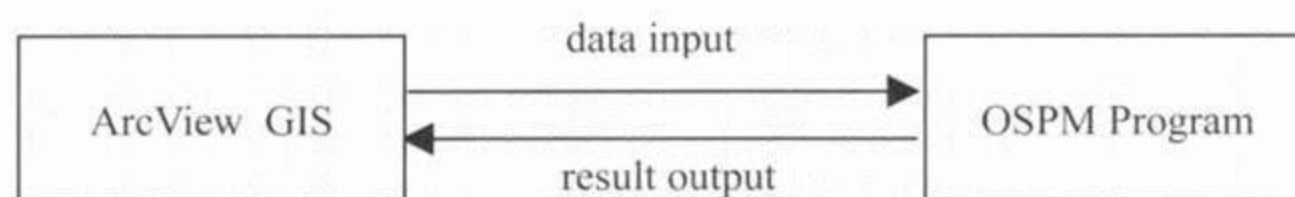


Figure 2 Process of integration OSPM in GIS

3. Pollutant Distribution

Hourly NO_x and CO concentrations accepted at roadside human height level were modeled under the normal condition of summer season in Macao. The classification of pollutant levels in the GIS coverage was according to national standard. As tourism industry is an exclusive economical sector in Macao, fulfilling type 1 standard is the ultimate goal. The result in Fig. 3, 4 showed that all the studied NO_x values (represented by NO₂) and CO values were satisfied national type 1 requirements under the model situation.

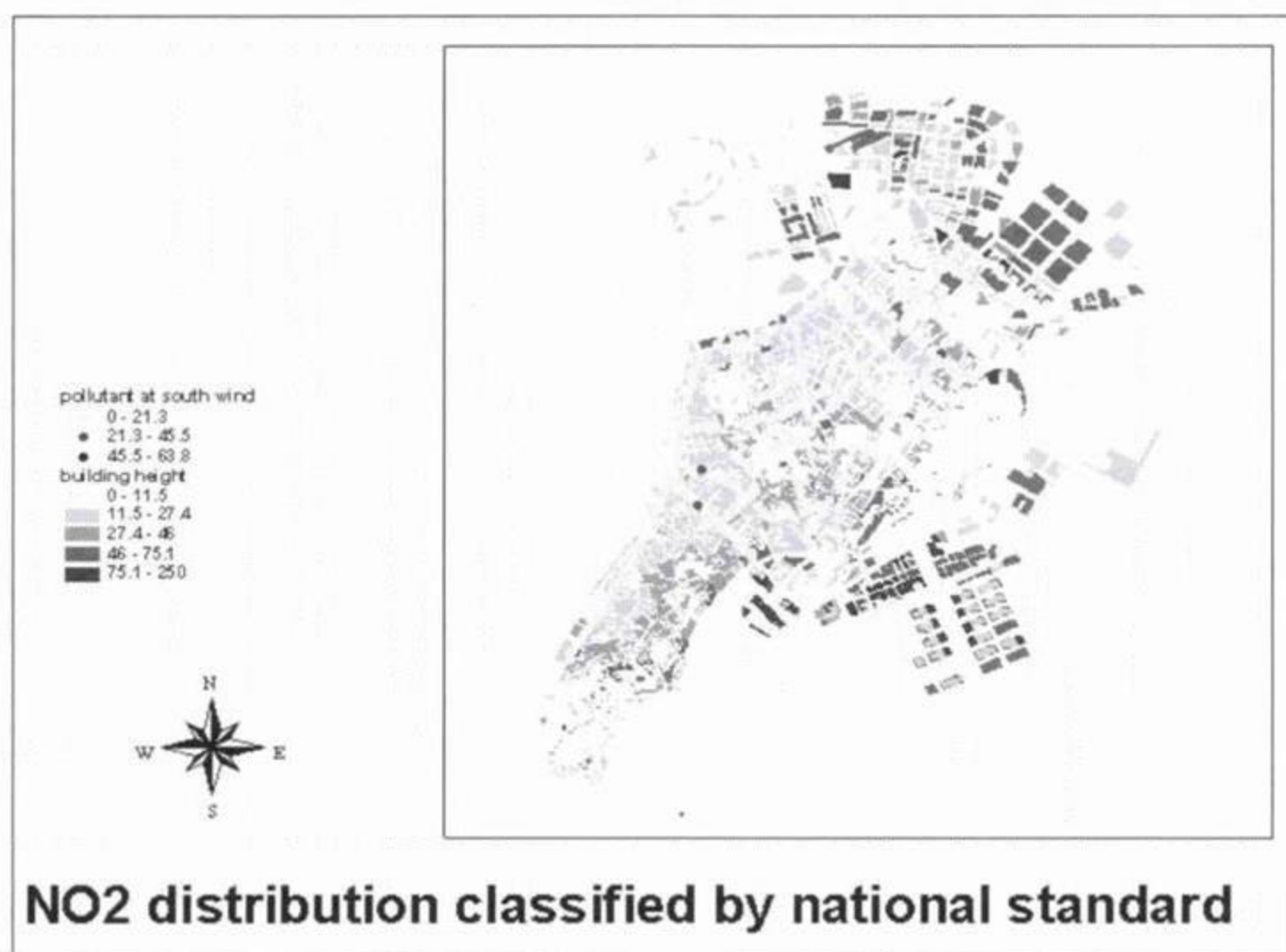


Figure 3 NOx classified by national standard, south wind direction

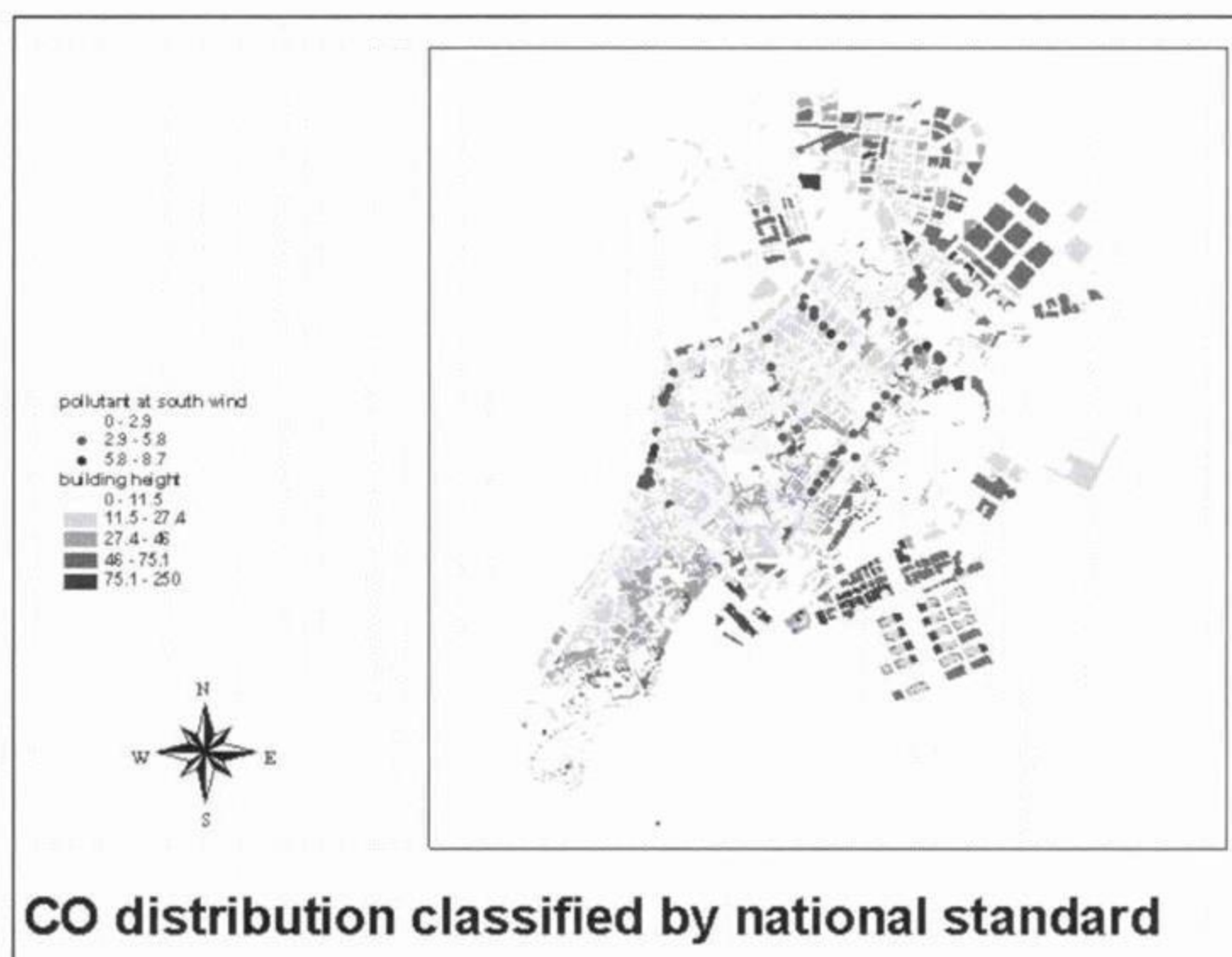
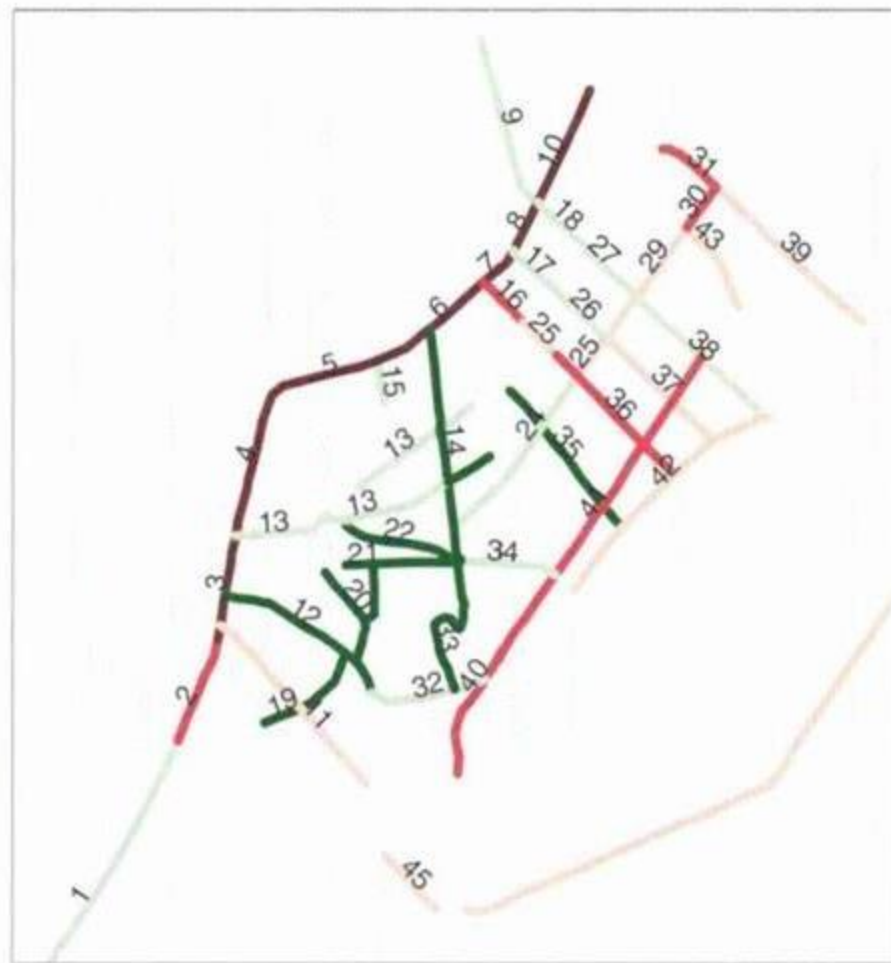


Figure 4 CO classified by national standard, south wind direction

4. Analysis of Geographic Influence on Air Quality

Although traffic flow is an important factor as it dominates the source of pollutant, the highest pollutant zones did not lay within the highest traffic flow areas as shown in Figure 6. Different levels of total traffic flows were distinguished by the color and the elevation. Coincided with what we commonly familiar with, the main street coded (Fig. 5) as 41, 38, 16, 25 and 3 to 10 serve a large amount of circulation while the central of peninsula is in low traffic flow intensity. However, this central area had a higher NOx in general.

The reason could be discovered by Fig.8. In order to examine distribution of the street canyon characteristic, H:W ratio (Height of building : Width of street) was displayed in front of each building block. Higher ratio implemented more serious of the street canyon effect and more difficult to disperse traffic pollutant. Five levels were classified in this figure using the natural-breaks method. An interesting finding was that the lowest H:W ratio existed round the boarder such as the west coast while the highest H:W ratio distributed at the central of peninsula. Hence, NO_x concentration was higher around the old district at the center of peninsula canyon with low traffic loading.



street code no & traffic flow

Figure 5 Total traffic flow comparison

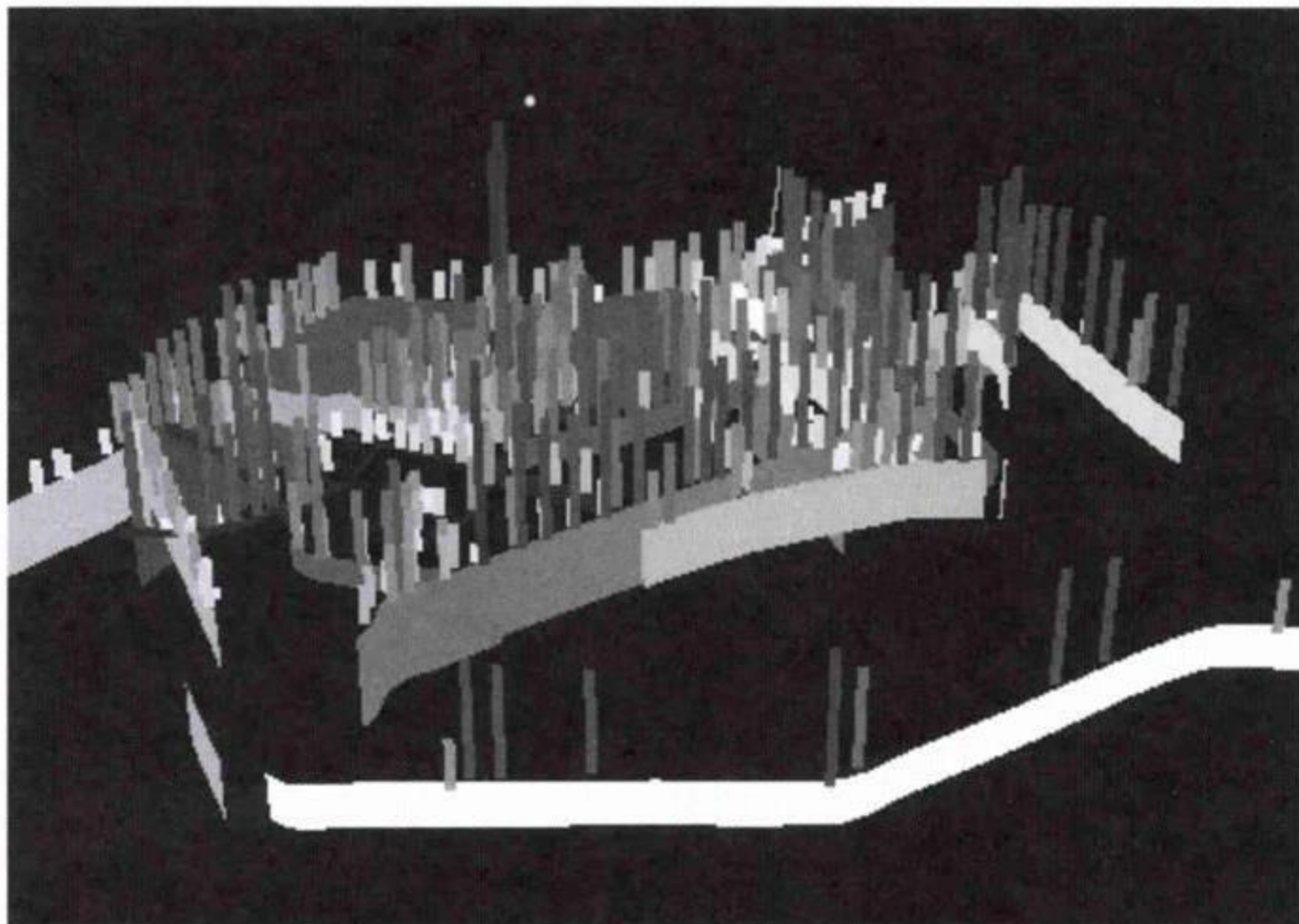


Figure 6 A 3D view of NO_x distribution vs. traffic flow

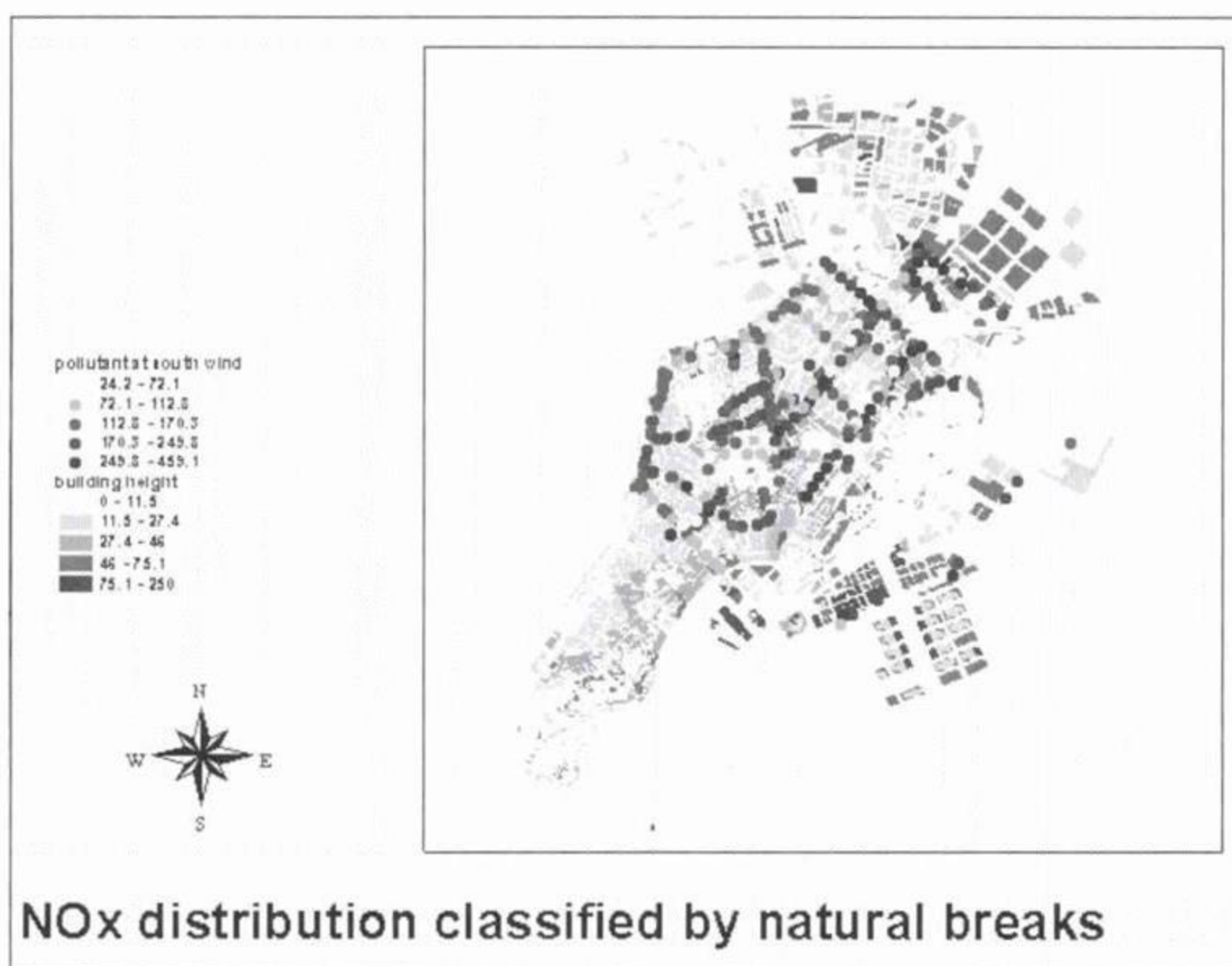


Figure 7 NOx concentration distribution

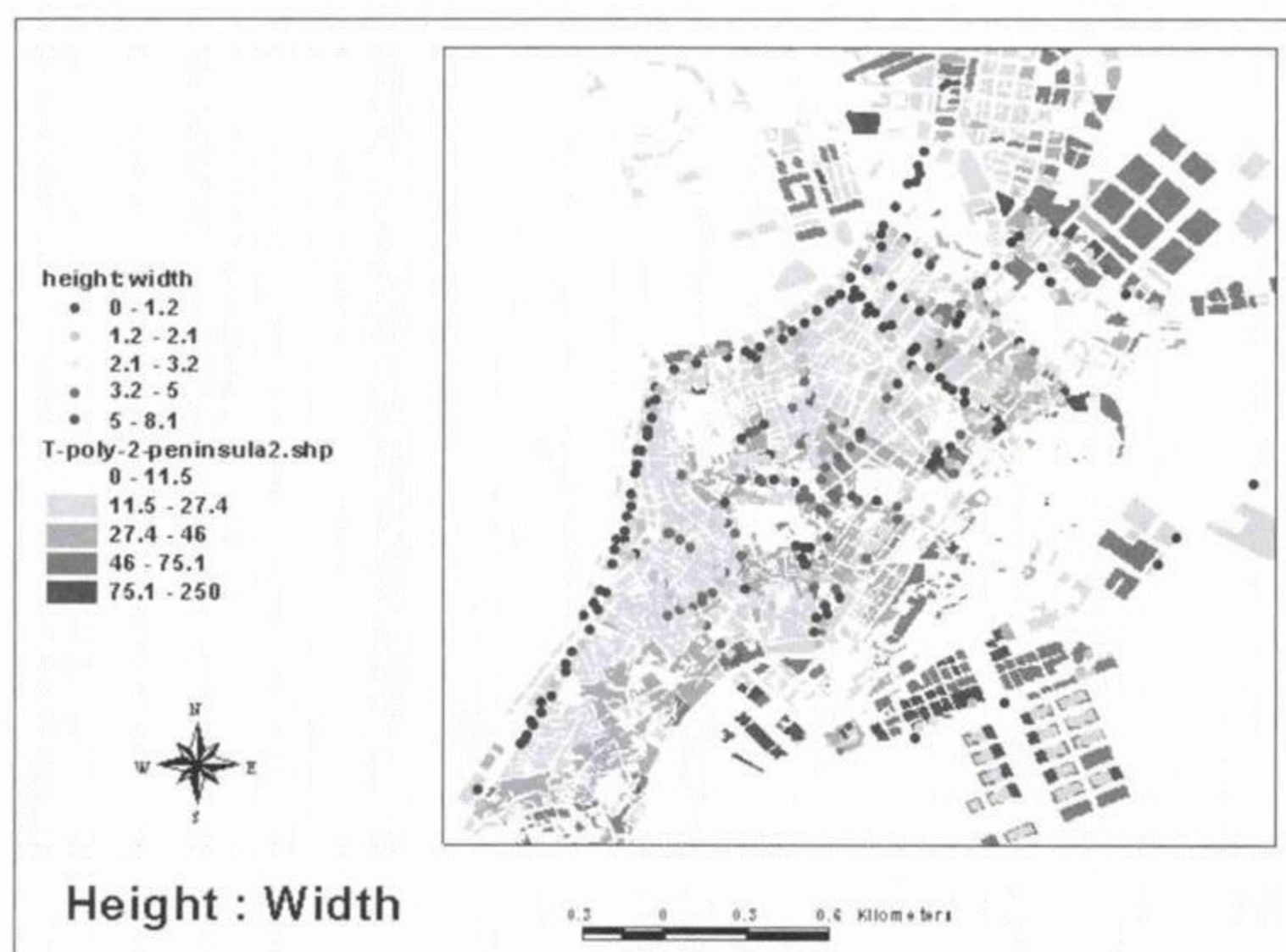


Figure 8 Intensity of street canyon

The finding of the roadside air quality highly related to the urban spatial characteristics has a significant meaning for the substantial urban development of Macao peninsula.

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Microorganism Control in HVAC System Using UV Light Under Two Different Working Conditions

L. M. Tam*, P. K. Wong*, S. C. Tam*

*Faculty of Science and Technology, University of Macau

K. T. Sou[†],

[†] Instituto Para os Assuntos Cívicos e Municipais

Abstract: Microorganism, such as the common yeast and mold or the dangerous tuberculosis (TB) is in fact a potential health threat to the occupants working in the indoor environment. Nowadays, most of the people work in indoor environment with air conditioning system. If the air-handling unit (AHU) of the central air conditioning system is lacked of proper maintenance, due to the dark-moist condition inside and the dust-microorganism collected in the filter within the unit, microorganism will have a perfect condition to generate and duplicate in large amount. The microorganism will be transferred from AHU to the occupying zone through the air duct. Currently, the common methods used in microorganism control are (1) ventilation method and (2) filtration method. Both methods are high in cost. The use of ultra-violet light for microorganism control may be the most cost-effective way. This study experimentally investigates the usage of ultra violet light in microorganism control using an environmental chamber with a scale down AHU under no-fresh air and full-fresh air experimental conditions. Favorable conditions for the growth of microorganism are generated within the AHU and the chamber before the activation of the UV light. Air samples are collected under different experimental conditions in sampling agar dishes using an impact air sampler for each air change after the activation of the UV light. Total bacteria count (PCA) and yeast and mold count (PDA) are measured in terms of colony forming unit (CFU). The experimental results show that the microorganism reduction rate is a function of the intensity of the ultra violet light, the air velocity, and the temperature of the air. When the experimental condition is set as no-fresh air, the microorganism can be reduced to a very low and safe level after 4 to 5 recirculated air changes. When the experiments are conducted under full-fresh air conditions, the microorganism reduction rate can be as high as 70 (PDA) to 95% (PCA) for low air velocity. In general, as indicated by the experimental data, the utilization of UV light for microorganism control is effective.

Keywords: Microorganism; HVAC; Ultraviolet light; Indoor air quality

Microorganism Control in HVAC System Using UV Light

Under Two Different Working Conditions

1. introduction

In Macau, high-rise buildings have developed rapidly in recent years and central air conditioning systems are widely used in such buildings. However, heating, ventilation, and air conditioning (HVAC) systems have been shown to act as a collection source for a variety of contaminants which may potentially affect health. Examples of such contaminants include mold, fungi, bacteria, and very small particles of dust and debris. For energy saving purposes, the HVAC designers prefer to use small amount of ventilation air and most of the supply air to the occupying zone are recirculated. As a result, concentration of the contamination will be increased gradually since fresh air is not enough to dilute the concentration of contamination inside the air conditioning area.

Buildings with an unusual numbers of occupants having physical problems have come to be described as having "Sick Building Symptoms". "Sick Building Syndrome" is the name commonly used to describe such energy efficient buildings that are not equipped with proper air quality control systems to remove indoor air contamination. The presence of microorganisms in indoor environments may cause allergic building-related illnesses. According to ASHRAE Handbook Fundamental (1990), some microorganisms such as bacteria, virus, mold under certain conditions may produce volatile chemicals that are malodorous or irritating, thus contributing to the development of what is called sick building syndrome. So such sick building syndrome (SBS) is an ailment of the 1990's in such tight design modern buildings. Harmful contamination in the air inside the buildings always lead to an increased number of complaints of headaches, eye and throat irritation, breathing difficulties, stuffy noses, bacterial infection, and even the spreading of diseases within hospitals, causing harm to the patients and occupants. Table I shows the common building-related diseases.

The environment inside the modern air ducting systems or air handling units provides sites for microorganisms to colonize and breed. The HVAC systems have been shown to act as a collection source for a variety of contaminates such as mold, fungi, bacteria, and very small particles of dust and debris that have the potential to affect health. The moist components of the central air conditioning system, such as filters, cooling coil, drain pan and drainage piping are ideal growing sites for bacteria development. The bacteria developed at these ideals growing sites will be distributed to the occupants and recirculated again inside the closed air conditioning area. Without an effective air quality control method, the density of the bacteria may increase day by day.

According to, Jagjit Singh (1996), Karin (1997), Morey and Elliot (1998), Rask et. al. (1998), and Jones (1999), their works indicate that the control of microorganism plays an important role in having good indoor air quality. Upper-room UV is an effective way in controlling microorganism as indicated by Riley et. al. (1976). However, according to Nardell (1997), research on upper-room UV to control microorganism is relatively inactive in decades. Firmly established application guidelines are not yet available and this method can only eliminate the microorganism in the room without cleaning the "source", i.e., the central air conditioning system. Since there is no systematic research on the application of ultra violet germicidal irradiation (UVGI) on central air conditioning system, this research hopefully can shed some light in this area.

The objectives of this study are to conduct a series of control experiments to investigate: (a) the influence of UVGI light intensity on microorganism control, (b) the influence of air velocity inside the air handling unit on microorganism control, (c) the influence of the UVGI lamp installation location on microorganism control. The total bacteria count (PCA) and yeast and mold count (PDA) obtained from air samples are used for analysis.

2. experimental setup

The experiments were conducted in a dynamic environmental chamber which is 3.1m width, 2m depth and 1.9m height (see figure 1). The chamber is connected to a computer linked air conditioning system with supply air volume control, temperature control, humidifying and de-humidifying functions, to generate various experimental conditions. Properties such as the dry bulb and wet bulb temperatures are monitored in different locations within the air conditioning unit. In order to simulate the central air handling system, supply and return air plenums with UVGI lamps inside are installed inside the chamber and are connected to the air conditioner with 250 mm diameter aluminum flexible duct to form a closed air-recirculation system. Two sets of additional fan inside the supply air plenum are used to generate different air velocity. The air velocity passing through the UVGI lamps is set to 2m/s or 4m/s for all the experimental runs. As seen in figure 2, an extra data logger is installed to monitor the dry and wet bulb temperatures across the UVGI lamp. The conditions of the test chamber are also monitor by the extra data logger.

table 1 Building-Related Diseases

Disease	History and Physical Examination	Laboratory Testing	Linkage	Causes
Rhinitis Sinusitis	Stuffy/runny nose, post-nasal drip, pale or erythematous mucos	Anterior and posterior rhinomanometry, acoustic rhinometry, nasal lavage, biopsy, rhinos copy, RAST or skin prick testing	Immunology skin prick or RAST testing, bradketted physiology	Direct occupational exposures; molds in the workplace; specific occupational factors (laser toners, carbonless copy paper, cleaning agents), secondary occupational exposures; pet(e.g. cat) danders brought from home.
Asthma	Coughing, wheezing, episodic dyspnea, wheezing on examination, chest tightness, temporal pattern at work	Spirometry before and after work on Monday, peak expiratory flow diary, methacholine challenge	Immunologic: skin prick or RAST testing. Physiologic: related to work*	See rhinitis/sinusitis
Hypersensitivity pneumonitis	Cough, dyspnea, myalgia, weakness, rales, clubbing, feverishness	DLCO, FVC, TLC, CKR, lung biopsy	Immunologic: IgG ab to agents present, challenge testing Physiologic (in acute forms): spirometry, DLCO	Molds, moisture
Organic dust toxic syndrome	Cough, dyspnea, chest tightness, feverishness	DLCO, TLC, WBC	Temporal pattern related to work	Gram negative bacteria
Contact dermatitis(allergic)	Dry skin, itching, scaling skin	Scaling rash, eczema biopsy	Patch testing	Molds, carbonless copy paper, laser toners
Contact urticaria	Hives	Inspection biopsy	Provocative testing	Office products(carbonless copy paper)
Eye irritation	Eye itching, irritation, dryness	Tear-film break-up time, conjunctival staining(fluorescein)	Temporal pattern	Low relative humidity, volatile organic compounds (allergic conjunctivitis), particulates
Nasal irritation	Stuffy, congested nose, rhinitis	Acoustic rhinometry, posterior and anterior rhinomanometry, nasal lavage, nasal biopsy	Temporal pattern	Low relative humidity, volatile organic compounds (allergic conjunctivitis), particulates
Central nervous system symptoms	Headache, fatigue, irritability, difficulty concentration	Neuropsychological testing	Temporal pattern(epidemiology)	Volatile organic compounds, noise, lighting, work stress, carbon monoxide, cytokines from bioaerosol exposure
Legionnaire's Disease	Pneumonic illness	History, Legionella culture from biopsy fluids	1)Organism isolated from patient and source. 2)immunologic watch	Aerosols from contaminated water sources, shower heads, water faucet aerators, humidifiers at home and at work, potable water sources (hot water heaters, etc.)

*(1) 10% decrement in FEV1 across workday,

FVC = forced vital capacity

(2) Peak flow changes suggestive of work relatedness, and

TLC = total lung capacity

(3) Methacholine reactivity resolving after six weeks away from

exposure

RAST = radio allergen sorbent test

DLCO = single breath carbon monoxide diffusing capacity

CXR = chest x-ray

IgG = class G immune globulins

FEV1 = forced expiratory volume in the first second

Dirty filters were used to generate microorganism inside the chamber. Since filter is acted as a collector of dust and microorganism, with suitable conditions, it may be treated as the source and it may reflect the actual working condition of an air handling unit. To generate the microorganism inside the test chamber, two dirty air filters (size: 1115 mmx655mmx19mm) from an air handling unit of a specific building were taken out and were placed inside the chamber for 48 hours. According to ASHRAE Standard 62a-89 (1990), the relative humidity in the test chamber is always set higher than 70% such that microorganism contamination can duplicate easily. Usually, microorganisms will grow up to around 10000 to 30000 CFU/m³ before the activation of the UVGI lamps.

The UVGI lamps used in this study is "SE161V0-16" by "STERIL-AIRE". The power rating for each lamp is 60W. Two sets of UVGI lamps are installed in the supply and return air plenum respectively with a combine power rating of 240W.

In order to standardize the experimental conditions within the chamber, the temperature is set to 21°C and the humidity is set to 50% RH for each experimental run. The air velocity passing the UVGI lamp is fixed to 2m/s or 4m/s. The UVGI lamps are turned on at the supply or (and) return air plenum with different power rating for different experimental runs.

3. results and discussions

Control experiments are conducted to show the relation of the bacteria reduction rate with the air velocity, the air volume passing through the UVGI lamp, the intensity (i.e. the power rating) of the UVGI lamp(s), and the initial condition of the microorganism under full fresh air and full recirculated air experimental conditions.



figure 1. dynamic environmental chamber

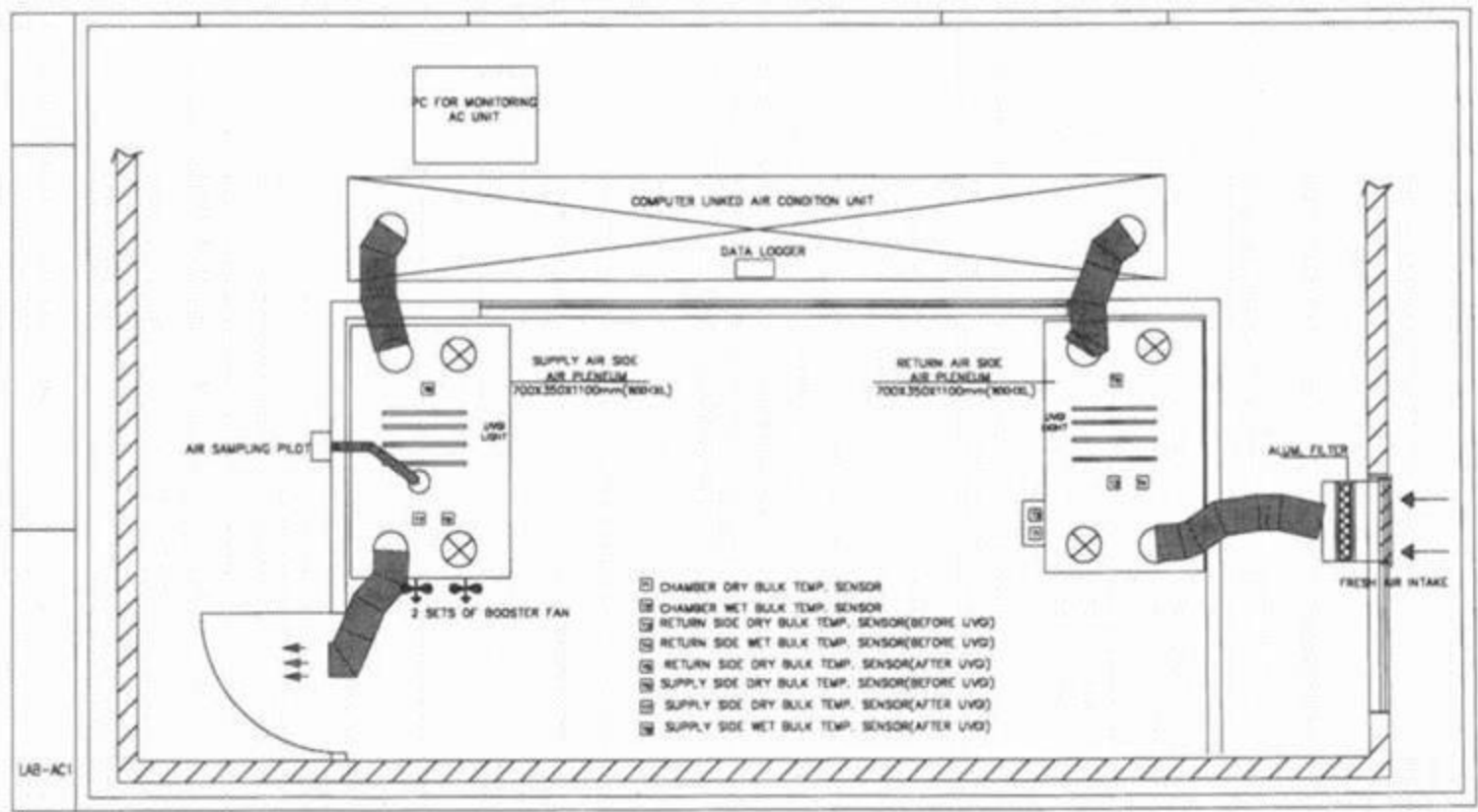


figure 2. schematic diagram for the dynamic environmental chamber

3.1. relation between microorganism reduction rate and different UVGI intensities under no fresh air condition

The installation of UVGI lamps will add extra cooling load on the air handling unit hence wasted energy. In order to control microorganism and save energy, it is necessary to investigate the relation between UVGI intensity in terms of the lamp's power rating and the reduction rate of microorganism. The ratio between the initial CFU values (before the activation of the UVGI lamp) and the CFU values after the activation of the UVGI lamp is used to show the reduction rate of microorganism as a function of different UVGI intensity (60w and 120w) for two sets of PCA data under 2m/s air velocity. As seen in figure 3, the reduction rate of microorganism is more effective for the 120w setting in the first few recirculated air changes. After that, the effect of higher intensity is not as significant. It can be concluded that high UVGI intensity can be set inside the air handling unit for the first few air changes. When the number of air change increases, it is recommended to switch to lower UVGI intensity for energy saving purposes.

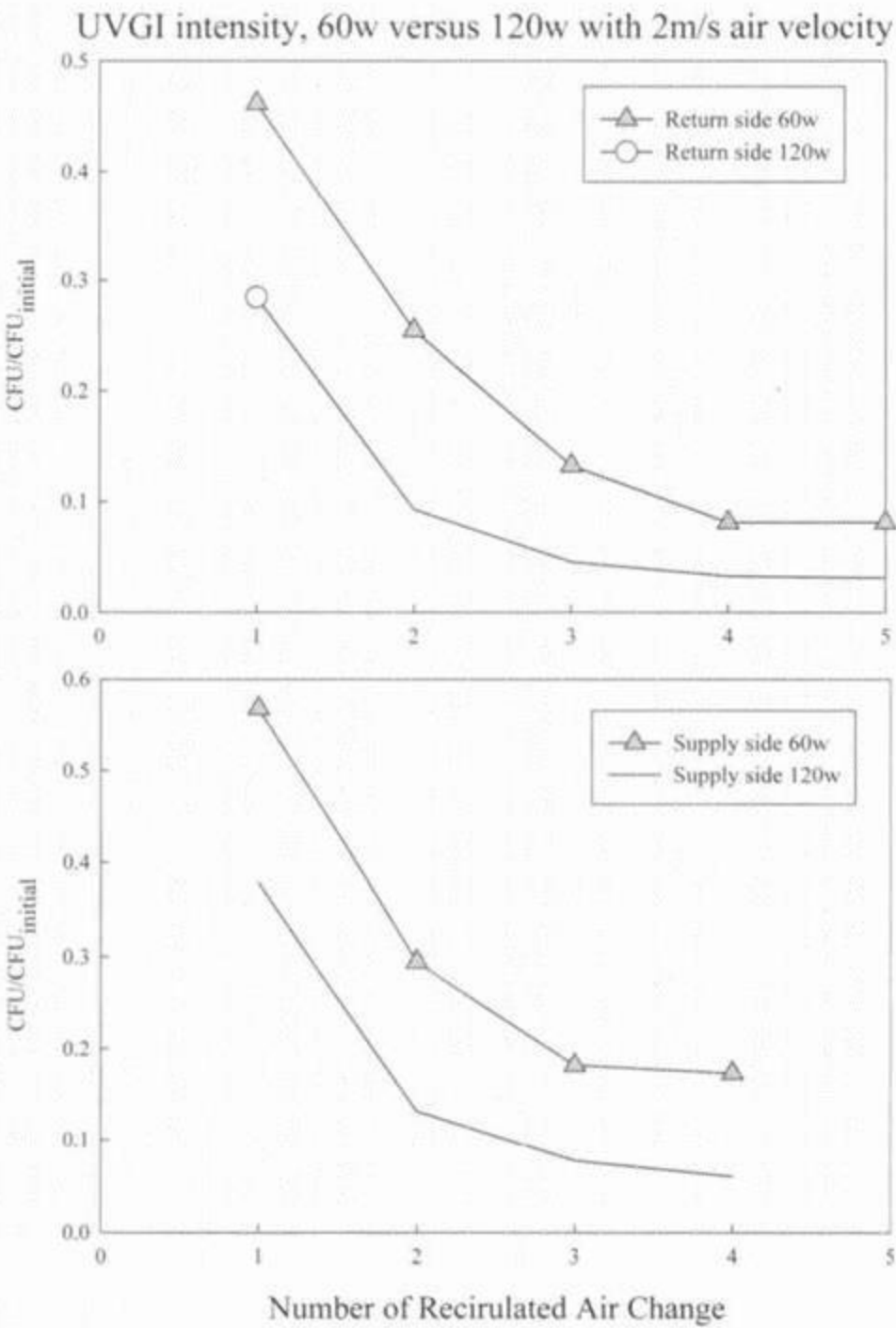


figure 3. effect of UVGI intensity on the reduction rate of microorganism.

3.2. relation between microorganism reduction rate and different UVGI installation locations under no fresh air condition

In order to evaluate the proper location for the installation of the UVGI lamp, experiments are conducted with the UVGI lamp installed in either the supply or the return air plenum. The CFU per 100 liters of air for both PCA and PDA counts were checked. As indicted by figure 4, it can be seen that the reduction rate of microorganism is higher when the UVGI lamp is installed in the return air plenum. The efficiency of the reduction rate of microorganism increases as air temperature increases since the temperature of the return air plenum is several degrees higher than the supply air plenum.

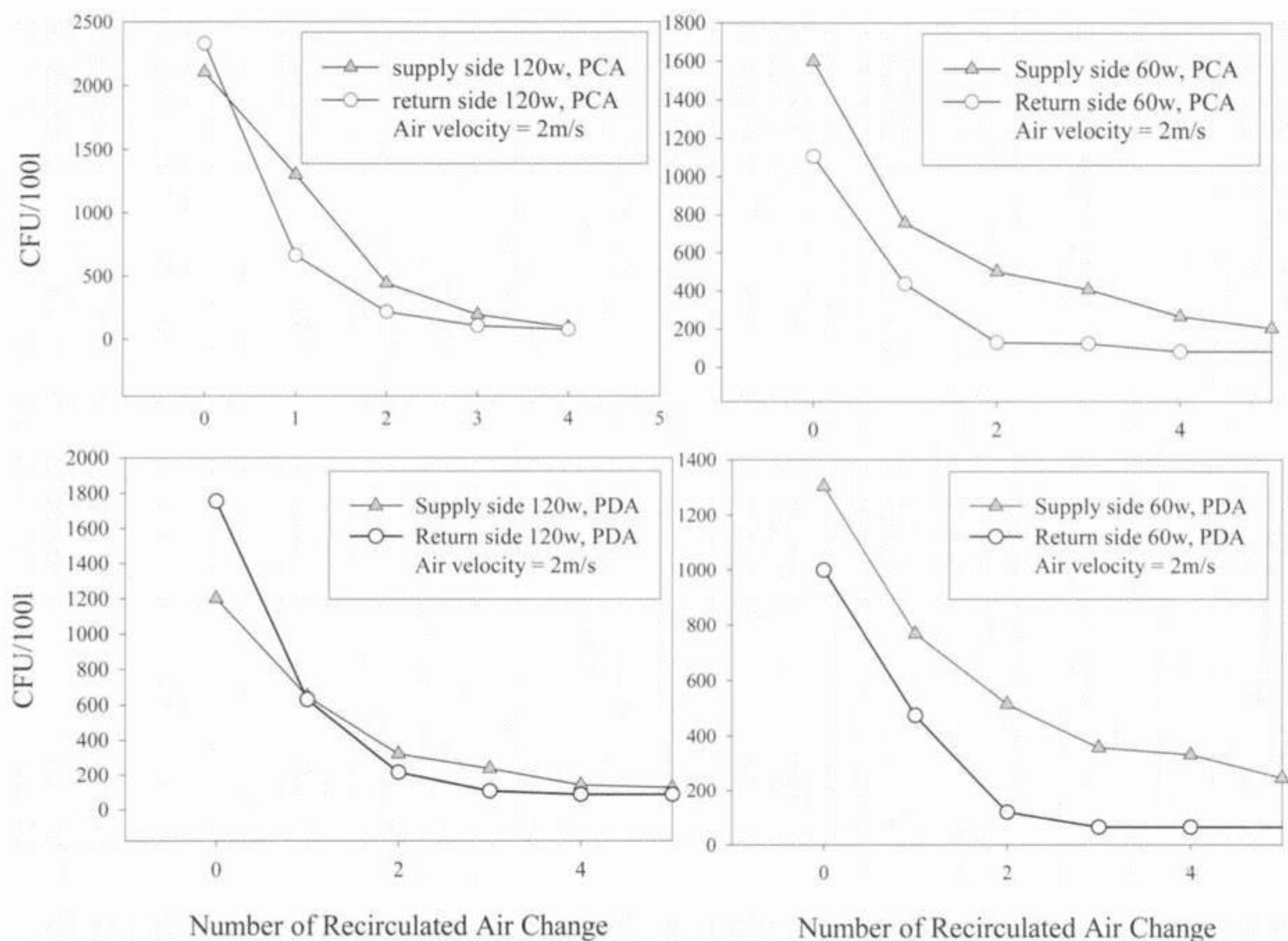


figure 4. effect of different UVGI installation locations on the CFU count per 100 liters of air

3.3. relation between microorganism reduction rate and different velocity and initial contamination conditions under no fresh air conditions

Two velocity settings (2m/s and 4m/s) inside the plenum were used to investigate the reduction rate of microorganism. These two velocities are commonly used in normal scale air handling unit. The results for PCA and PDA counts under different velocity were examined separately. The initial contamination condition for each experimental run was different. As mentioned in the previous section, it is very difficult to establish the same initial conditions for each experimental run and hence difficult to make comparisons. Therefore, the ratio between the initial CFU values (before the activation of the UVGI lamp) and the CFU values after the activation of the UVGI lamp is used to examine the data.

Figures 5 and 6 show the reduction rate of microorganism for PDA and PCA counts in terms of CFU ratio under different velocity and initial contamination conditions. For PDA, 13 experimental

runs with 65 data points were used for the 2m/s setting and 11 experimental runs with 82 data points were used for the 4m/s setting. For PCA, 13 experimental runs with 67 data points were used for 2m/s setting and 7 experimental runs with 50 data points were used for 4m/s setting.

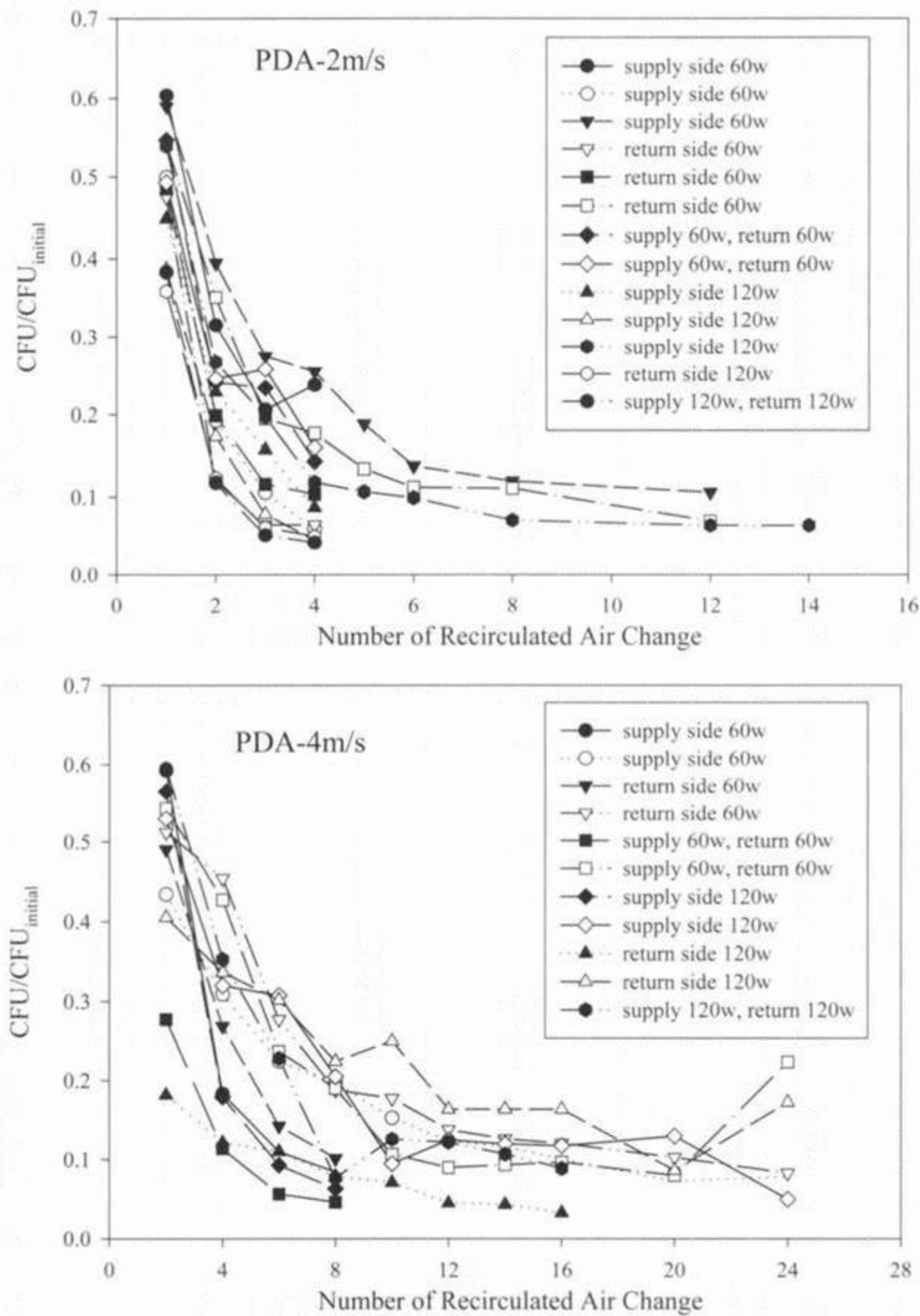


figure 5. PDA reduction rate under different velocities and initial contamination conditions

For the case of PDA, it can be seen that for the 2m/s air velocity setting, the CFU values drop to about 40 to 60% of the initial value (i.e. $CFU/CFU_{initial} = 0.4$ to 0.6) after the first recirculated air change. The sharp decreasing rate continues until the number of recirculated air change approximately equals to 4 to 6. It can be observed that the CFU ratio for different experimental runs approached a specific asymptote ranged from around 0.03 to 0.13. When the velocity is set to be 4m/s, it can be observed that the CFU values drop to about 20 to 60% of the initial values after the recirculated air change is equal to 2. It means that for the 4m/s velocity setting, it takes twice the time to achieve the killing rate of microorganism similar to the 2m/s setting. The CFU ratio decreases as the number of recirculated air change increases. When the number of recirculated air change approximately equals to 8 to 12, the CFU ratio value will reach asymptotic values ranged from 0.04 to 0.14 depends on different initial conditions.

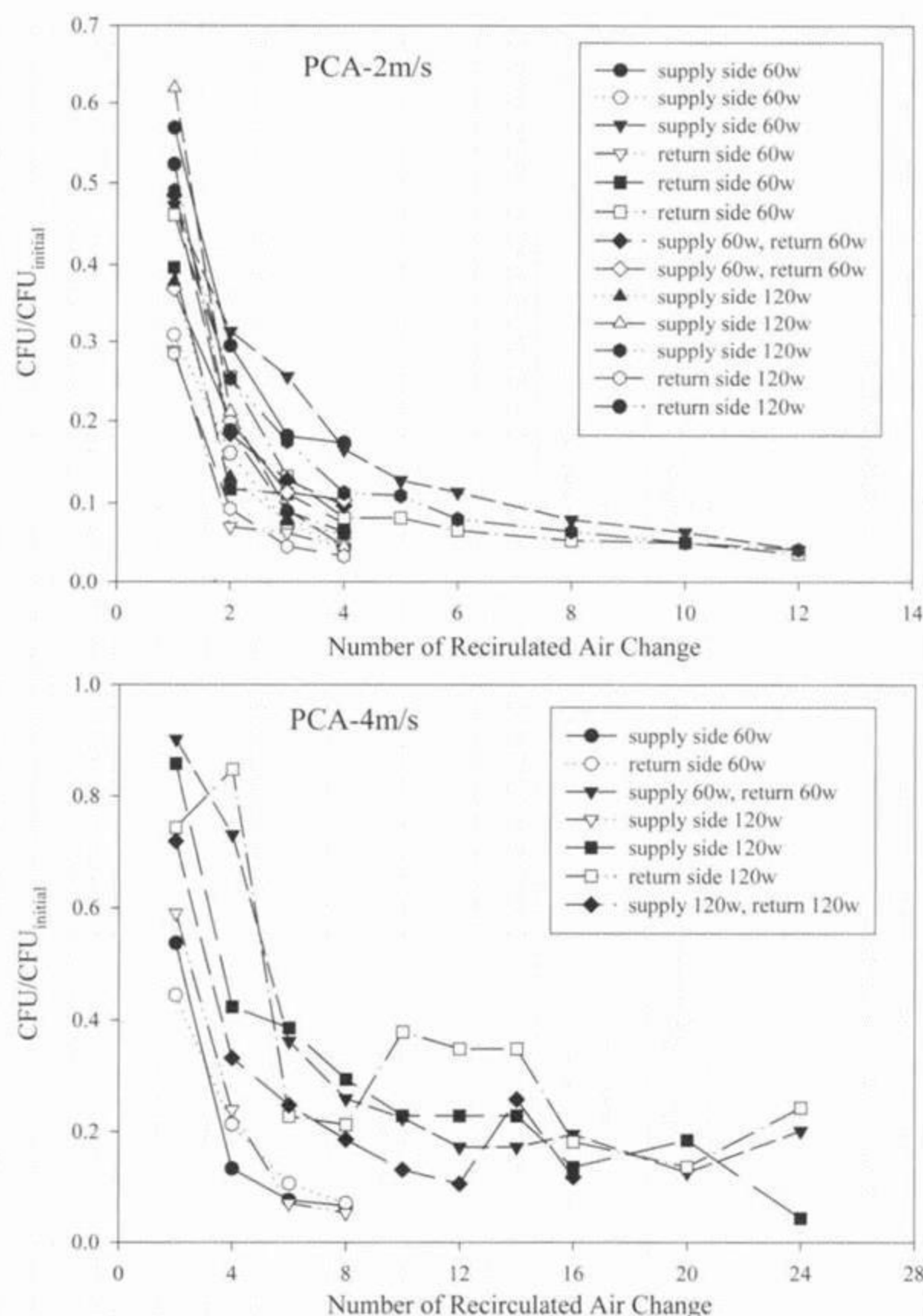


figure 6. PCA reduction rate under different velocities and initial contamination conditions

For the case of PCA, the trends are very similar. As seen in figure 6, the CFU values for the 2m/s setting drop to about 30 to 60% of the initial value after the first recirculated air change. The sharp decreasing rate continues until the number of recirculated air change approximately equals to 4 to 8. The asymptotic behavior is not as obvious as the previous case. When the air velocity is equal to 4m/s, it can be seen that the CFU values drop to about 40 to 90% of the initial values. When the number of recirculated air change increases, the CFU ratio decreases sharply. Similar to the case of PDA, it takes twice the time to achieve the comparable reduction rate when the velocity setting is equal to 4m/s.

3.4. the reduction of microorganism with long term UV light exposure with full recirculated air

It would be interested to see how the microorganism's reduction rate changes with time with the UV light keeps turning on for a relatively long period of time. The on time of the UV light ranges from 16 to 148 hours. Figure 7 showed the results. The reduction rate for PCA and PDA ranges from 64 to 100%. Therefore, turning on the UV light within the central air conditioning system continuously could keep the microorganism in a relatively low level.

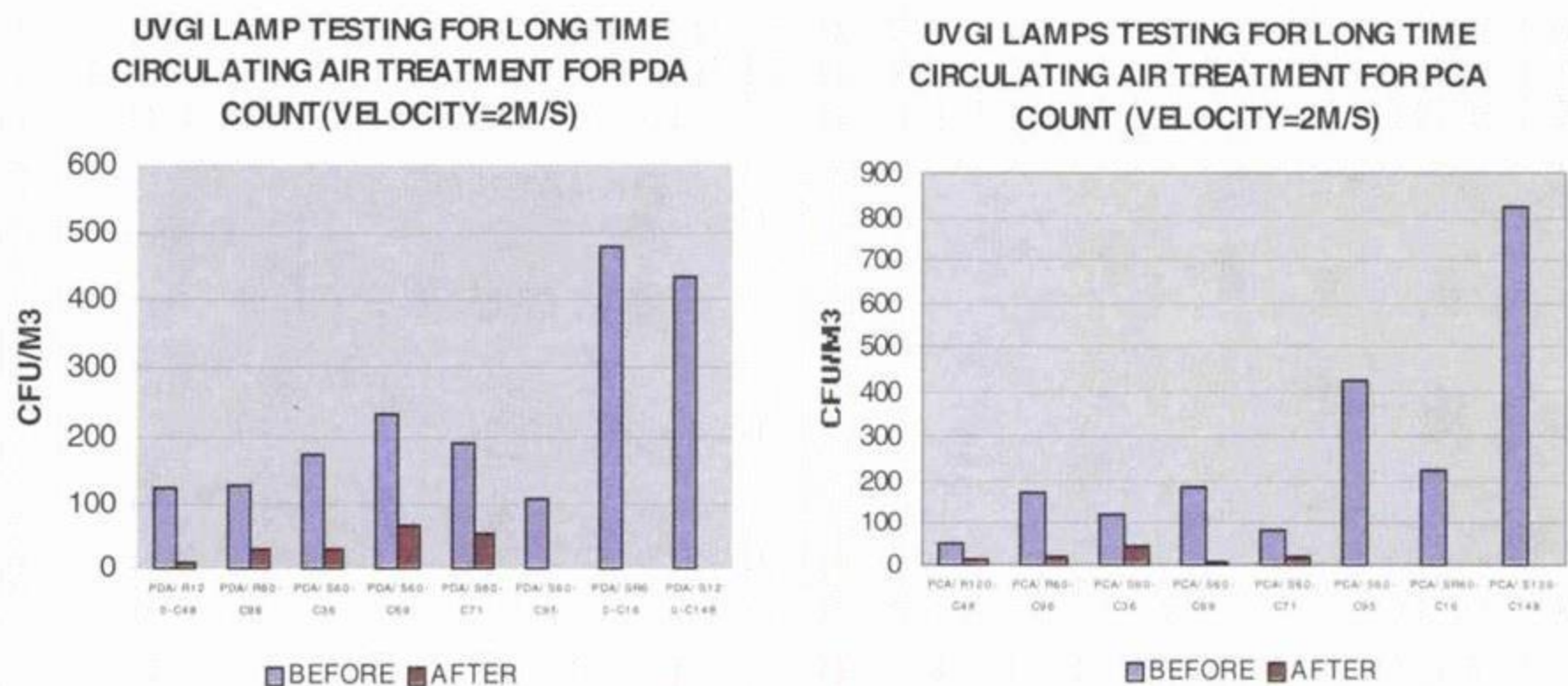


figure 7 the reduction of microorganism recirculated air under long term UV exposure

3.5. microorganism reduction rate under full fresh air condition

The microorganism reduction rate under full fresh air condition is investigated. Unlike the no fresh air condition, the air will only be treated by UV light once. The air velocities are set to be 2m/s and 4 m/s respectively. Figure 8, and 9 showed the results.

As seen in these figures, the microorganism reduction rate ranges from 50 to 95% for PCA and 32 to 70% for PDA when the air velocity is set to be 2m/s. When the air velocity is set to be 4m/s, the reduction rate ranges from 22 to 74% PCA and 10 to 64% for PDA. Since the time for the microorganism exposed to UV light is shorter for 4m/s, the reduction rate for 2m/s is higher than that for 4m/s in general.

4. conclusion and recommendation

Experiments are conducted to investigate the use of ultraviolet light to control the microorganism generated within the air handling unit. The results indicate that high UVGI intensity is very effective in reducing the microorganism for the first few air changes. When the number of recirculated air change increases, the effect is not that significant in comparison to low UVGI intensity. The results also indicate that it is better to install the UVGI lamp in the return air plenum since the killing rate of microorganism increases as temperature increases. The effects of velocity and initial CFU conditions on the killing rate of microorganism are also investigated. According to the experimental data, it takes twice the time for the high velocity (4m/s) setting to have approximately the same killing rate of microorganism as the low velocity setting (2m/s).

The microorganism reduction rate under 100% recirculated air with long term UV exposure and full fresh air working condition with the UV light installed in the plenum are also investigated. For long term exposure, the microorganism reduction rate with the UV light turned on for 16 to 148 hours, the microorganism can be kept in a relatively low level as indicated by the reduction rate. For one time UV light treatment, it can be seen the microorganism reduction rate is higher for 2m/s in comparison to 4m/s. This is directly due to the UV exposure time. One time treatment is therefore still effective for low air velocity. When the air velocity increases, the microorganism reduction rate decreases.

Since this experiments conducted in this study are based on the worse operating condition (100% recirculated air) and the best operating condition (100% fresh air) of the air handling unit, to make this study more comprehensives, similar experiments should be conducted with fresh air added into the recirculated air stream.

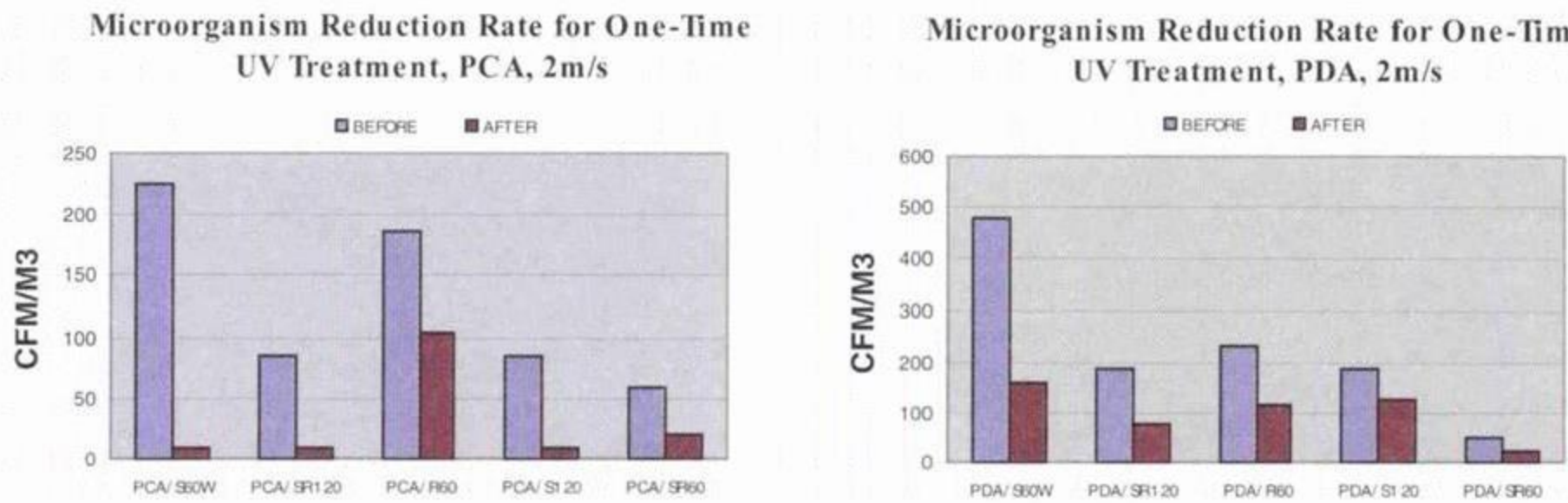


figure 8 one-time UV treatment for PCA and PDA for 2m/s air velocity

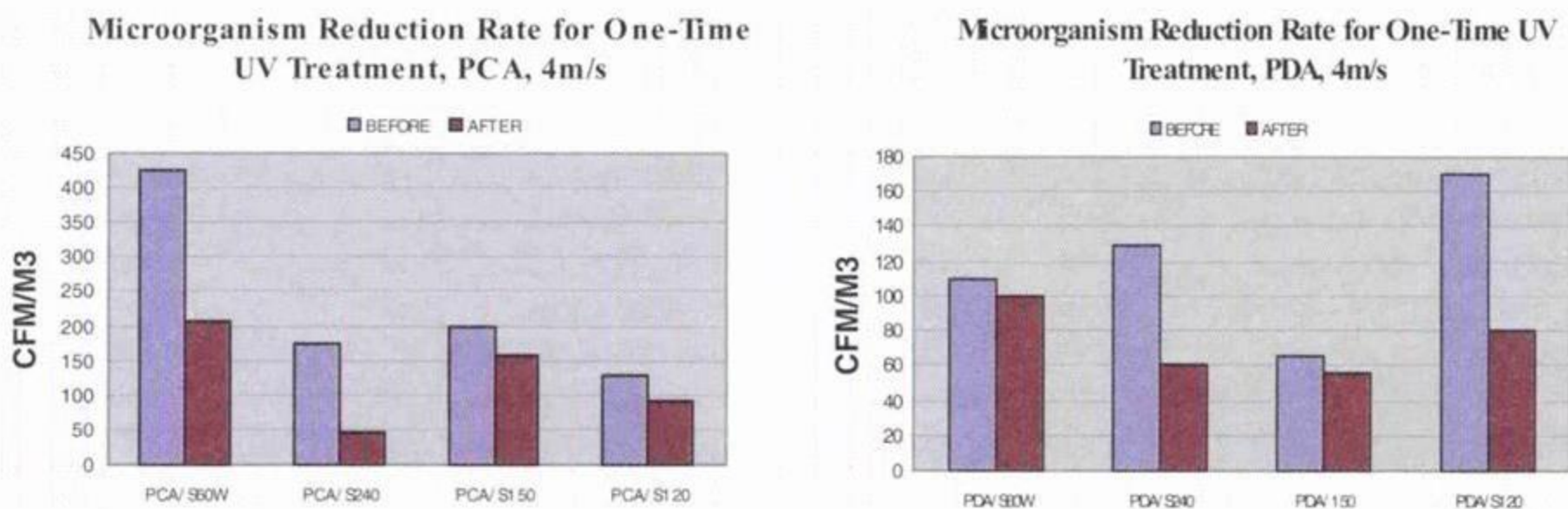


figure 9 one-time UV treatment for PCA and PDA for 4m/s air velocity

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Development of an Air-Quality Index Forecasting System for Macau – A Memoir

K. M. Mok

Department of Civil and Environmental Engineering

Faculty of Science and Technology, University of Macau

PO Box 3001, Macau SAR, China

Abstract: The rapid population and economic growth in the Pearl River Delta and the local traffic volume increase during the recent years has imposed stresses on the atmospheric environment as well as the well being of Macao residents. The increase of public awareness on the ambient air quality propels the development of Macau air quality research studies. The initial study was focused on the concentrations of sulfur dioxide alone with the goal of developing an accurate meteorological dispersion model. Due to insufficient background information and the small geographic size of Macau, this approach was found to be uneconomical or feasible at the time. Application of artificial intelligent techniques in short-term air quality forecasting is then considered and found that it may satisfy the immediate needs of Macau. Researches in this direction were carried out since 1997 and successful application results in Macau are found. The success of the neural network applications on air quality forecasting attracted cooperation intentions from academic and industrial units, and an international EUREKA project was materialized and approved in 1998. The project is with acronym INTELAIR and number EU 1920. The present paper summarized some of the main results obtained in these areas and a memoir on the development of an intelligent air-quality index forecasting system for Macau, which is adopted by the Macau Meteorological and Geophysical Bureau (SMG) for their forecasting applications in Macau's daily AQI public broadcast, is presented.

Keywords: Macau; Air Quality Index; Artificial Intelligence; EUREKA INTELAIR

1. Introduction

Macau comprises a Macau peninsula and two islands of Taipa and Coloane with a total area of 25.8 km² today. It lies on the west side of the Pearl River Delta located at the southeast of China. Hong Kong and Guangzhou situate at 60 km to its East-NorthEast and 105 km to its North, respectively (figure 1). With such geographical location, Macau experienced rapid economic growth as its neighbors in the Delta area in the last decay. The estimated year-end population in 1997 was 422,000^[1] which is a 16 % growth from the 363,784 population size in 1991^[2]. The corresponding increase of annual gross domestic product (GDP) within that period is from 32,118 million MOP^[2] in 1991 to 58,472.1 million MOP^[1] in 1997, which translates to a 82 % growth. The consequence of growth is the increase in energy consumption hence, the decrease of environmental quality, especially the ambient air quality. Recognizing the potential impacts of the regional growth on the ambient air environment, the aspiration to comprehend the air-concentration condition as well as its scientific modeling in Macau was stimulated. The year was 1997 and the rest is history.



Figure 1. Location of Macau

2. The Initial Stage

At the beginning of the air-quality study started in 1997, focus was put on the development of the more traditional mathematical meteorological and dispersion models, which can describe the relationship between pollutant emission, transmission and ambient air concentrations of the air pollutant as a function of space and time. Feasibility study of this approach was carried out first. It was identified that the calculation quality of the said models depends on precise information on the pollutant emission, wind field, topographic details and even the individual turbulence condition of the atmosphere, just to name a few. Immediately, it was found that there was no systematic study on pollutant emission inventory in Macau. Therefore, the essential local emission data required by the models is not well defined. In addition, the air quality of Macau could also be influenced by the conditions of its neighboring region, e.g. southern China, through long-range transport,^[3] which puts in additional difficulty in obtaining accurate estimation of pollutant emission. Even the pollutant emission could be estimated with sufficient accuracy, if the models developed intend to include both of the local and foreign effects from the closer emission sources in the southern China, the spatial scales of such simulation models could range from a few kilometers to tens of kilometers. Then the small size of Macau would appear in the simulated area more or less as a point. With such concern as well as the ill-defined emission inventory of Macau, development of the traditional dispersion model was decided as non-economical (if only the situation in Macau is concerned) or feasible at the time.

3. Re-defining the Goals from the Actual Needs of Macau

Knowing that the development of a traditional air-quality model for Macau may not be suitable at the time, a step backward was taken to review the overall condition in Macau so that research and development goals could be redefined and actual immediate needs of Macau could be satisfied. The first step was to identify what reliable information was available and what were the immediate needs of Macau.

It was found that the most reliable information available for Macau is the monitored ambient air quality, which has been officially recorded by the Macau Meteorological and Geophysical Bureau (SMG) since 1987. During the period of 1987 to 1991, a total of ten measuring stations at eleven locations (figure 2) in Macau were set up to monitor the concentration of seven pollutants including sulfur dioxide, black smoke, nitrogen dioxide, lead, sedimentary particulate, total suspended particulate with diameter less than 100 μm , and PM-10. The SMG had been releasing air quality reports annually since then and the smallest time scale of the released pollutant concentrations is 24 hours; i.e. daily averaged pollutant concentration could be obtained. At the same time, it was also learned that the SMG was starting to set up three continuous ambient air concentration monitoring

stations to accurately and effectively record the air quality and planned to launch an air-quality-index release scheme for public information in 1999. Therefore, it was identified that the development of a computationally economical and intelligent short-term air-quality-forecasting system would meet the immediate needs of Macau and subsequent researches were set in this direction.

4. Neural Networks for Air Quality Forecasting

The main objective in air quality forecasting is to predict the values of future air concentration based on values of a number of input variables such as the past air concentration record, related meteorology factors and/or the pollutant emission rate. This is basically a regression problem with the goal of finding a suitable function, which maps the air concentration to related factors. Artificial neural networks (ANN) were considered in vogue for these kinds of modeling applications due to their computation efficiency. A pilot study in testing the applicability of ANN in air quality forecasting for Macau was launched immediately in 1997.

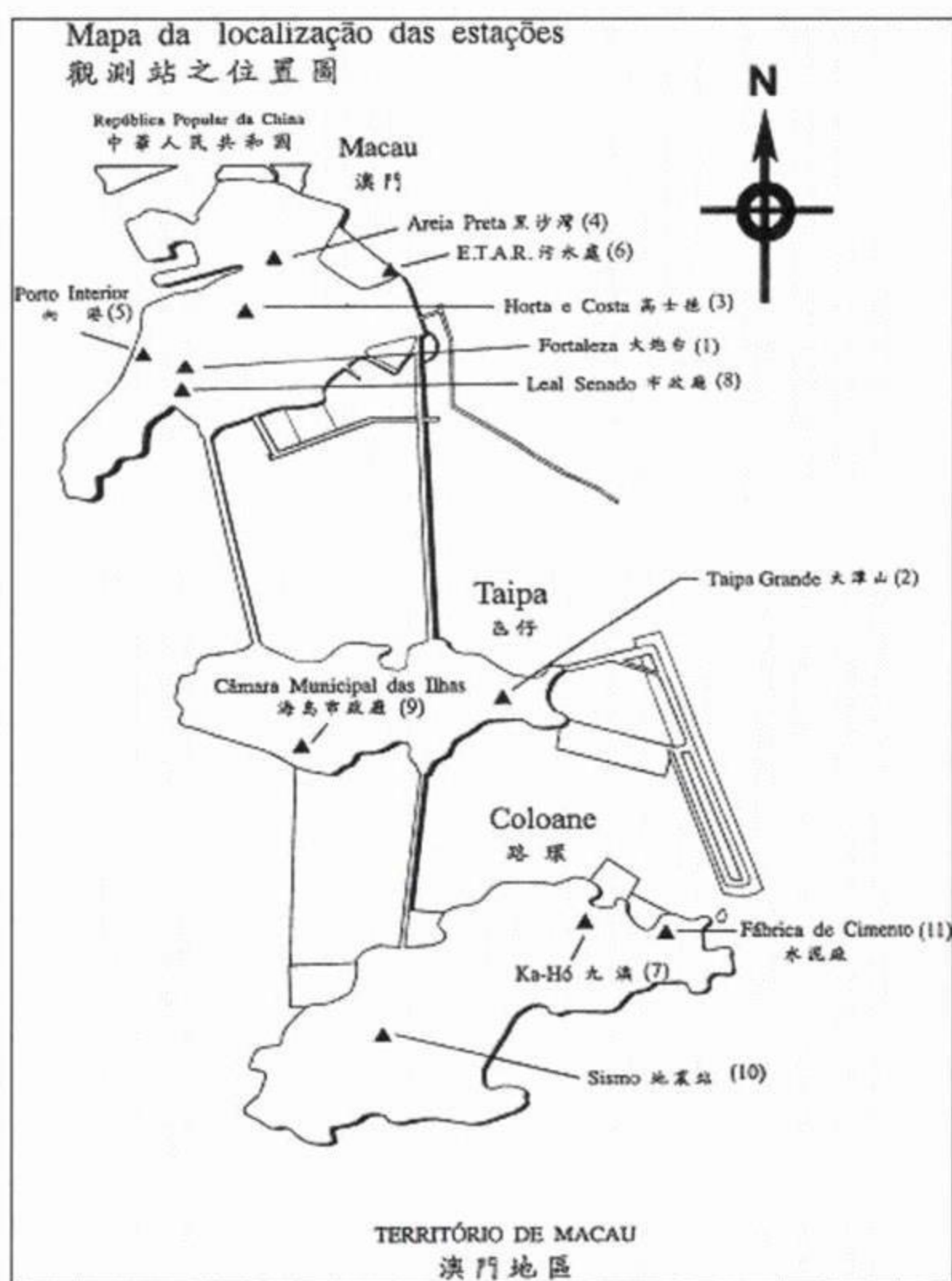


Figure 2. Air-quality monitoring stations in Macau (Source: SMG)

The pilot study was focused on the application of artificial neural networks in short-term prediction of sulfur dioxide concentration in Macau. The reason for selecting sulfur dioxide was due to the more complete data collection that could be obtained from the SMG at that time. In addition sulfur dioxide is one of the primary pollutants, which is of great concern to Macau, as well as to many urban cities in the world, due to its contribution to the local acid rain problems. The forecasting model was based on the assumption that the effects of the underlying deterministic forces like the changing meteorological factors is implicitly embedded in the changing air-concentration time series and the future value of air concentration is only a function of its past records; i.e.

$$C_t = F(C_{t-\tau_1}, C_{t-\tau_2}, \dots, C_{t-\tau_d}) \quad (1)$$

where C_t is the air concentration value at time t , τ_k with $k = 1, 2, \dots, d$, is a non-negative increasing finite sequence and d is called the embedding dimension and F is an unknown function relating the air concentration value at t with its past record. Note that F is a nonlinear function, which could be estimated by ANN through training. The ANN used in the study was a three-layered feed-forward network of the back-propagation type and a definition sketch of it with n inputs, r neurons, and one output is shown in figure 3.

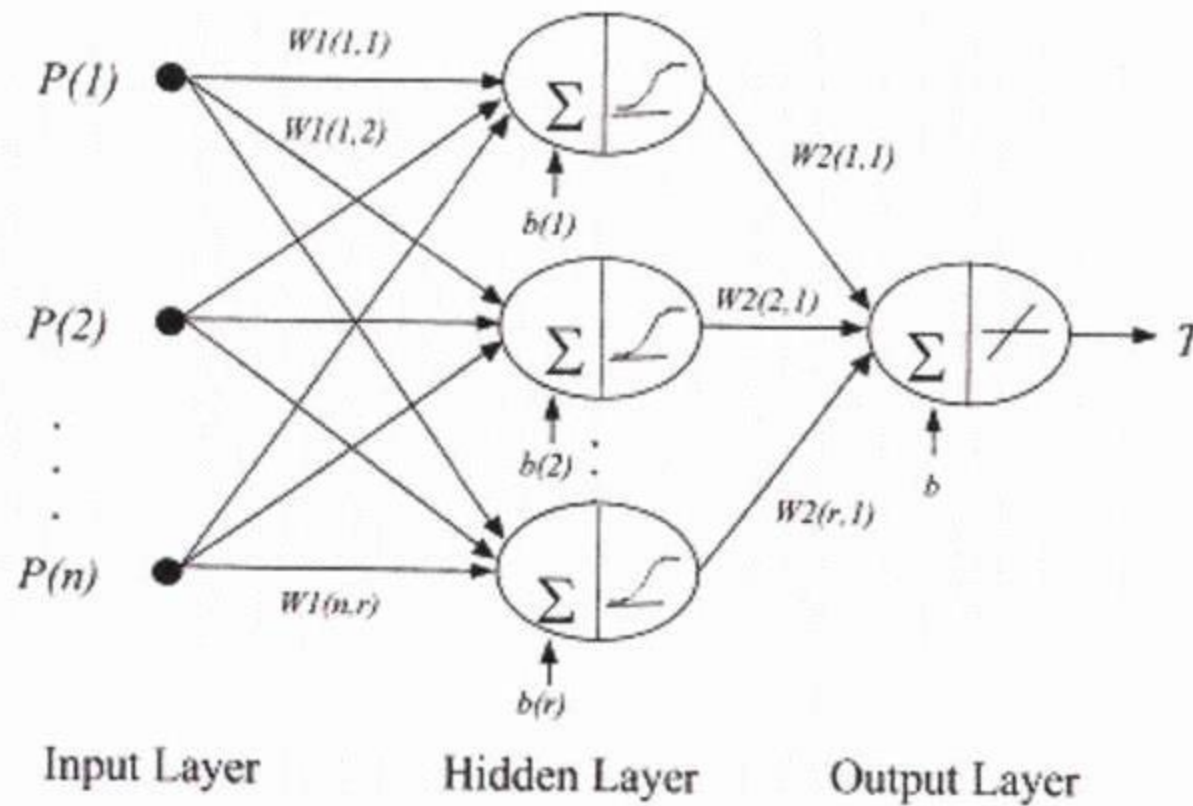


Figure 3. Definition sketch of a three-layered feed-forward network

In the input layer, pattern $P(1), P(2), \dots, P(n)$ is taken. Then they are linearly transformed by the neurons of the input layer. The output signal generated is weighted with $W1(n, r)$ and fed to each of the r neurons of the hidden layer. Then each of the neurons of the hidden layer sums all the weighted input signals and bias $b(r)$ and produces an overall unit activation which is then converted to an output signal by utilizing a sigmoid transfer function. They are then weighted with $W2(r, 1)$ and fed to the neurons of the output layer. At last the weighted input signals and the bias b of the output layer are summed to produce a unit activation, which is linearly transformed to yield the calculated target value T . The learning method used is the well known back-propagation algorithm.^[4] Briefly, this method lets the network learn from the examples sequentially and the values of the adaptive parameters of the networks, the weights and the bias, will be updated so that the cost function and the sum-squared error can be stochastically minimized.

Study of the daily averaged sulfur dioxide (SO_2) concentrations in 1995 indicated that *Areia Preta* at the northern part of Macau was the area that had the highest potential of being polluted; it was identified as the representing site for investigation and the data recorded in the period of October to December was used. In that study, it was assumed that the future SO_2 concentration is only related to the evolution patterns occurred during the past thirty days. Based on this assumption and the size of the selected SO_2 data, two separate tests were set to examine the prediction ability of the ANN model. One test was to use the data of October to train the networks to predict the first five-day SO_2 concentrations of November (Case I), and the other test used the November data to predict the first five-day values of December (Case II). For these five-day ahead prediction tests, the ANN shown in figure 3 was adopted with $n = 3$ and a time lag of 4 days; i.e. to predict the SO_2 concentration at day t , the daily averaged values at days $t - 4$, $t - 8$, and $t - 12$ were used. This arrangement was based on a trial and error basis and it was found suitable for both cases. However, the numbers of neurons used in the hidden layer for both cases were slightly different with Case I using 9 and Case II using 12. The results showed that the accuracy of the ANN model was within 14.5% and 13.7% for the two testing periods, respectively. Figure 4 shows the results of the Case II. These promising results indicated that the ANN could be used to develop efficient air-quality prediction models. The ideas were immediately accepted by the research community and an article "Short-Term Prediction of SO_2 Concentration in Macau with Artificial Neural Networks" was published in an international journal in 1998.^[5]

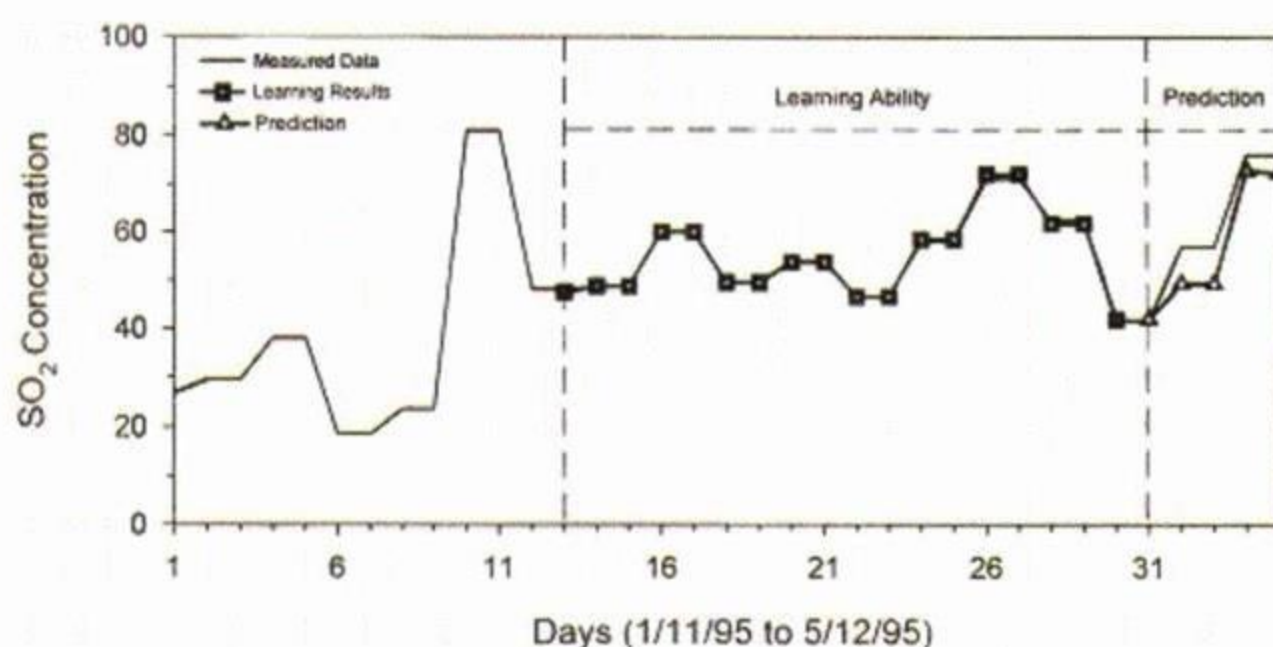


Figure 4. ANN calculated and measured daily average SO_2 concentration at A. Preta of Macau during the period of November 1 to December 5 of 1995; the units for SO_2 concentration is in $\mu\text{g}/\text{m}^3$.

5. International Cooperation - EUREKA

The success of the neural network application on air quality forecasting attracted cooperation intentions from academic and industrial units, and an international EUREKA project was materialized and approved in 1998. The project was with acronym INTELAIR and number E! 1920. The title of the project was "Hybrid System for Urban Air Pollution Management: Artificial Intelligent and Classical Modeling". It officially started in January 1999 with a total budget of 0.5 million Euro. It was a two-year project with seven partners; W. S. Atkins Portugal, Consultores e Proj. Internacion. Lda. and IDAD - Instituto do Ambiente e Desenvolvimento/Environment were from Portugal, United Kingdom Meteorological Office - Consultant Group was from the United Kingdom, Northeast Electric Power Group Corporation of China and Peking University/Institute of Environmental Engineering were from Mainland China, University of Macau/Faculty of Science and Technology and Companhia de Electricidade de Macau, S.A.R.L. were from Macau. The project concluded in January of 2001.^[6] The objective of the INTELAIR project was to develop a hybrid air quality forecasting system, which uses both the traditional and artificial intelligent approaches. Our research team at the University of Macau was in charge of developing the INTELAIR Neural Component, the core of the intelligent system that could forecast future air quality based on historical information. The first workshop of the project was held in March of 1999 at University of Macau (figure 5).



Figure 5. Group picture of the partners in the first workshop held at University of Macau in March of 1999.

6. The Air-Quality Index Forecasting System for Macau

As mentioned in Section 3, the SMG installed three continuous monitoring stations (figure 6) in both the Macau peninsula and the Taipa Island so that the needs of air-quality information from the

concerned units and more important, the general publics, could be satisfied. In March of 1999 (the same week of the first INTELAIR workshop), the SMG introduced an air quality index (AQI) system and officially started to report the last 24-hour's AQI to the general publics; at the same time, the SMG also planned to release one-day ahead AQI forecasts in two years time, i.e. in March of 2001. This immediate local need confirmed our beliefs^[7]; i.e. an air-quality forecasting system was in demand. The parallel objectives of our INTELAIR project and the SMG's AQI plan propelled the development of an air quality index forecasting system for Macau within our group.

6.1. Validation of ANN Application in AQI Prediction

Before the development of the final forecasting system for Macau, application tests of ANN on the AQI prediction were carried out to re-ensure the approach is feasible even though previous application on short-term SO₂ concentration forecasting was successful.^[5] The validation was done by using data recorded at the Northern Zone air-quality monitoring station during 1999 for modeling and testing. It is noted that the Northern Zone station is one of the three stations (figure 6) with operation started in 1999. In the following subsections, an introduction to the AQI system used in Macau and the corresponding validation process are presented.

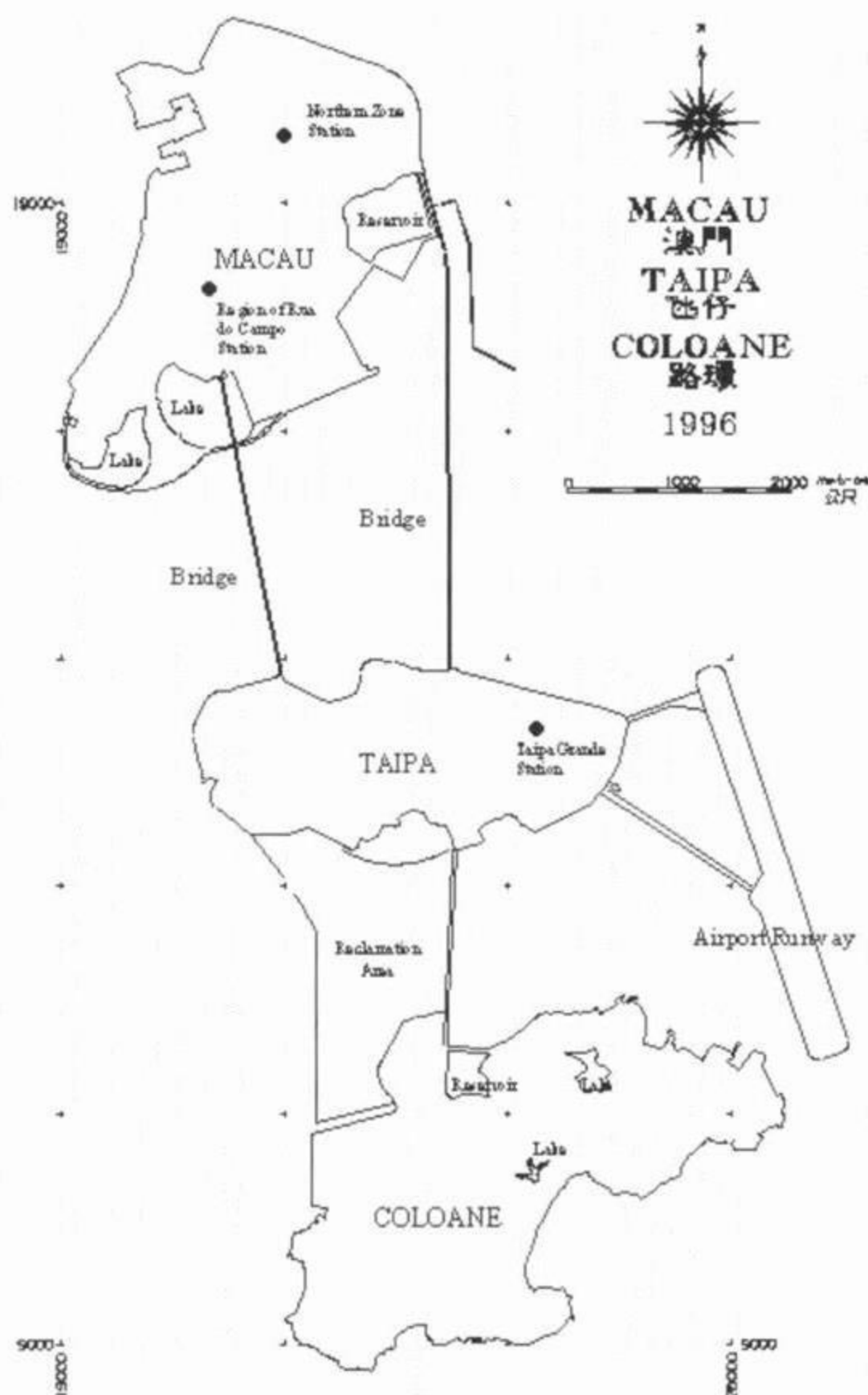


Figure 6. The three continuous air quality monitoring stations in Macau with operation started in 1999; • represents the station.

6.1.1. The Air Quality Index System of Macau

The SMG started in March 1999 to monitor and to report the last 24-hour air quality situation to the general public in Macau. The air quality is recorded through three continuous monitoring stations located at various sites in Macau as shown in figure 6. The Measured pollutants and the categories of Macau ambient air quality recorded at these stations are as listed in Table 1. It is noted that the operation of the Taipa Grande and the Northern Zone Stations was started in March 1999 while the Region of "Rua do Campo" station was started in July 1999. It is also noted that one more station was added in Taipa Island in 2001 so that a total of four stations are currently operating. In the present paper only the first three stations are introduced.

Table 1. Measured pollutants and site category of the three monitoring stations

Station Name	Category	Measured Pollutants
Taipa Grande	Ambient	PM10, SO ₂ , NO/NO ₂ /NO _x , O ₃
Northern Zone	High Residential Area	PM10, SO ₂ , NO/NO ₂ /NO _x , O ₃ , CO
Region of "Rua do Campo"	Roadside	PM10, NO/NO ₂ /NO _x , CO

In order to provide an easy understanding of air quality to the general public, the SMG uses an Air Quality Index (AQI) system, which classifies the air quality into six levels. Currently there are three daily AQIs announced by the SMG. Each of them represents different categories of the air quality; i.e. the general ambient air quality, the air quality at high (population density) residential area, and the roadside air quality. All of the Macau AQIs are daily indexes that range from 0 to 500 and they come from the linear-segmented transformation of at most five major pollutant concentrations. The major pollutants used are respirable suspended particulate (PM₁₀), sulphur dioxide (SO₂), nitrogen dioxide (NO₂), carbon monoxide (CO) and ozone (O₃). The range of AQI is classified into six bands by the SMG as listed in Table 2. The AQI value within different band corresponds to different level of air pollution. In general the higher the AQI value, the poorer the air quality. In calculating the announced daily AQI, separate sub-index is calculated for each of the five major pollutants. The sub-indexes of the pollutants are converted from their measured concentrations based on the scale listed in Table 3. The reported AQI is defined as the maximum value among this set of sub-indexes. The daily AQIs are reported everyday in the afternoon and the sub-indexes are calculated from corresponding concentrations taken during the period of 12:00 P.M. of the reporting day back to 12:00 P.M. (or 5:00 A.M.) of yesterday for the 24-hr (or 8-hr) average cases.

Table 2. AQI range and its associated air pollution level

AQI	Air pollution level
0 – 50	GOOD
51 – 100	MODERATE
101 – 200	BAD
201 – 300	VERY BAD
301 – 400	SEVERE
401 – 500	HARMFUL

Table 3. AQI conversion table used in Macau

Air Quality Index (AQI)	PM ₁₀ 24-hr average ($\mu\text{g}/\text{m}^3$)	SO ₂ 24-hr average ($\mu\text{g}/\text{m}^3$)	NO ₂ 24-hr average ($\mu\text{g}/\text{m}^3$)	CO 8-hr average ($\mu\text{g}/\text{m}^3$)	O ₃ 8-hr average ($\mu\text{g}/\text{m}^3$)
0	0	0	0	0	0
50	100	60	80	5000	80
100	150	150	150	10000	160
200	350	800	280	17000	350
300	420	1600	565	34000	600
400	500	2100	750	46000	800
500	600	2620	940	57000	1000

6.1.2. Application Results at the Northern Zone Station

In the validation study, the air pollutant concentrations at the Northern Zone Station recorded during the period of April 1 to July 31, 1999 were used. At this station, all of the five major pollutants used to determine the AQI were measured. Hence the procedures included the development of five independent ANN models for predicting the sub-indexes of the five pollutants. Then the maximum of the five predicted sub-indexes would form the predicted AQI. Since the data available were the daily air quality sub-indexes and the corresponding daily AQI recorded from April to July 1999, for model training, testing and performance evaluation, the data were divided into three segments. The data from April and May were for model training while the data in June were for testing and adjusting the ANN structure. The data in July were used for model performance evaluation by comparison with the model predictions.

As the validation study used the three layer feed-forward network as discussed in Section 4 for forecasting the air quality index one day ahead, the main structure elements needed to be determined were the input pattern (layer) and the number of neurons in the hidden layer. Nevertheless, the exact analysis of the ANN structure is still inconclusive and it is usually determined experimentally. Since the forecast of the final AQI was determined from the maximum of the five predicted sub-indexes, individual model for each pollutant was built as mentioned earlier. It was assumed that the variation of the next day air quality depended only on condition of no more than twelve of the past days (the input pattern). Further assumption was that the ANN structures of all the sub-index models were the same and they were determined by the overall performance on the final AQI prediction test through experiments.

Experimenting on the combinations of different input patterns and their corresponding numbers of neurons in the hidden layer, the most effective ANN structure with the least root mean square error on AQI in the test period was adopted for final prediction. It was found that the structures with the past three-day air quality information as inputs together with eight neurons in the hidden layer gave the best test results. The trained system was then used to forecast the AQI of July 1999 for model verification. Since the forecast was one day ahead, the input data were updated with the actual measurements for each new forecast as it advanced through the month of July. The forecasting results comparing with the actual measurements are shown in figure 7. It was shown that the Models performed relatively well with a mean absolute percentage error of only 15.8% in the prediction period and 96.8 % of the forecasts give the correct main contribution pollutant. It was noted that the air quality index variation in July was relatively mild except at the last ten days; in that period the concentrations experienced a mild jump. The models were still able to capture the main trend with relatively good predictions. The tests were successful and the results were published in Air Pollution VIII^[8] in the summer of 2000.

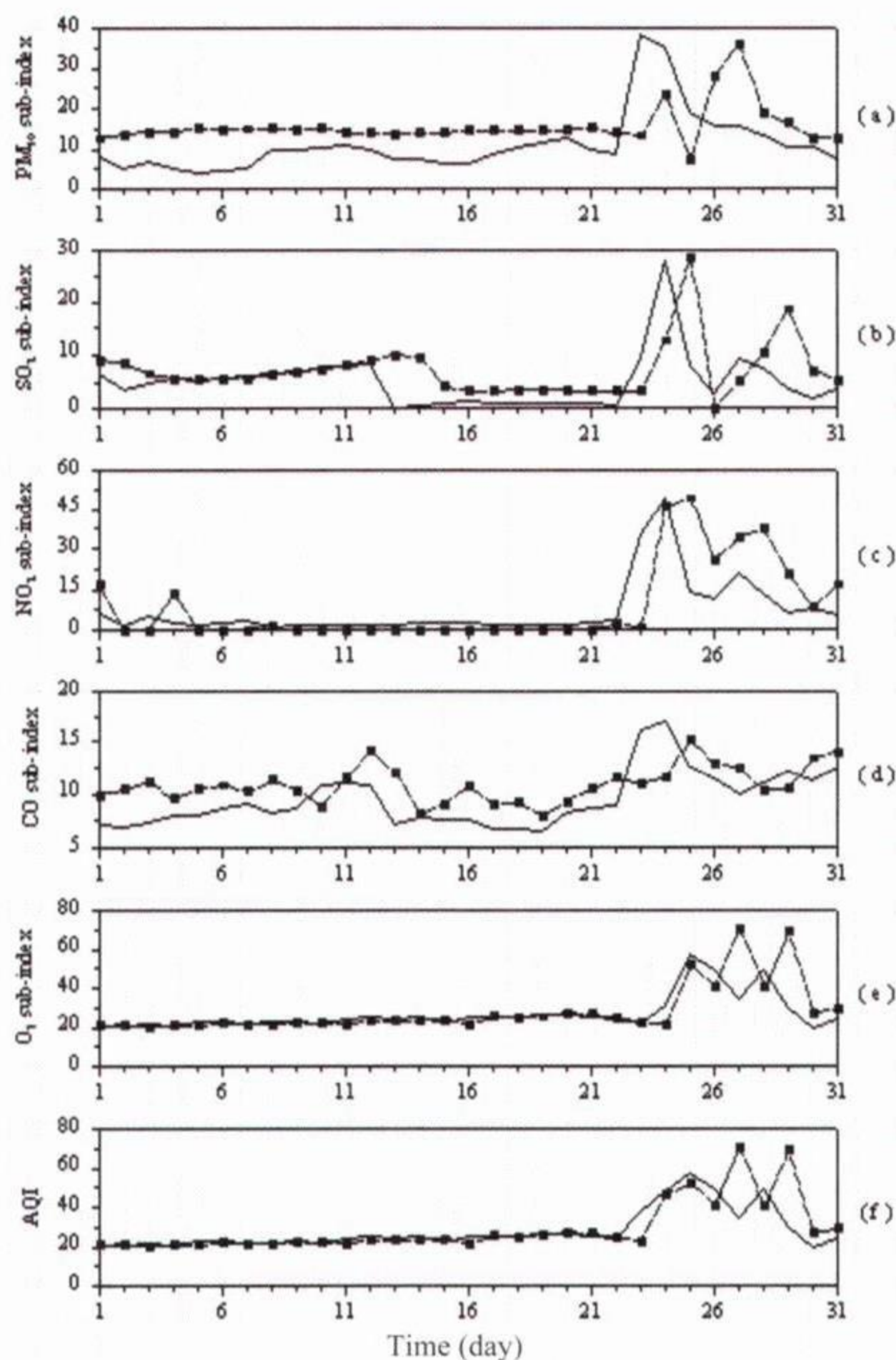


Figure 7. Comparison between the forecasted daily pollutant sub-indexes and final air quality index (AQI) with the measurements at the Northern Zone Station during July of 1999; AQI (graph (f)) represents the maximum value within all pollutant sub-indexes (graphs (a) to (e)) of the corresponding day; — is measurement and - - - is prediction.

6.2. The INTELAIR Neural Component

With the feasibility study of the ANN application on Macau AQI forecasting completed successfully, development of the intelligent forecasting system for Macau was fully carried out in later part of the year 2000. When the system development was carried out, the objectives of the project were determined first. It was decided that the system would be application software, which is intended to be used by a wide range of professionals with different backgrounds. The development objectives were the software system must be able to run on easily accessible personal computers operated with the most popular operation system; the Microsoft Windows. The developed software should then be a Windows based, user-friendly yet efficient system, and the user should not be required to obtain other commercial software to have it run properly. It was then decided to program the system with the Microsoft Visual Basic 6.0. The complete system includes two stand-alone units: a database

management system mainly for air quality information management and analysis (figure 8), and a neural unit for air-quality forecasting bearing the name “INTELAIR Neural Component” (figure 9).

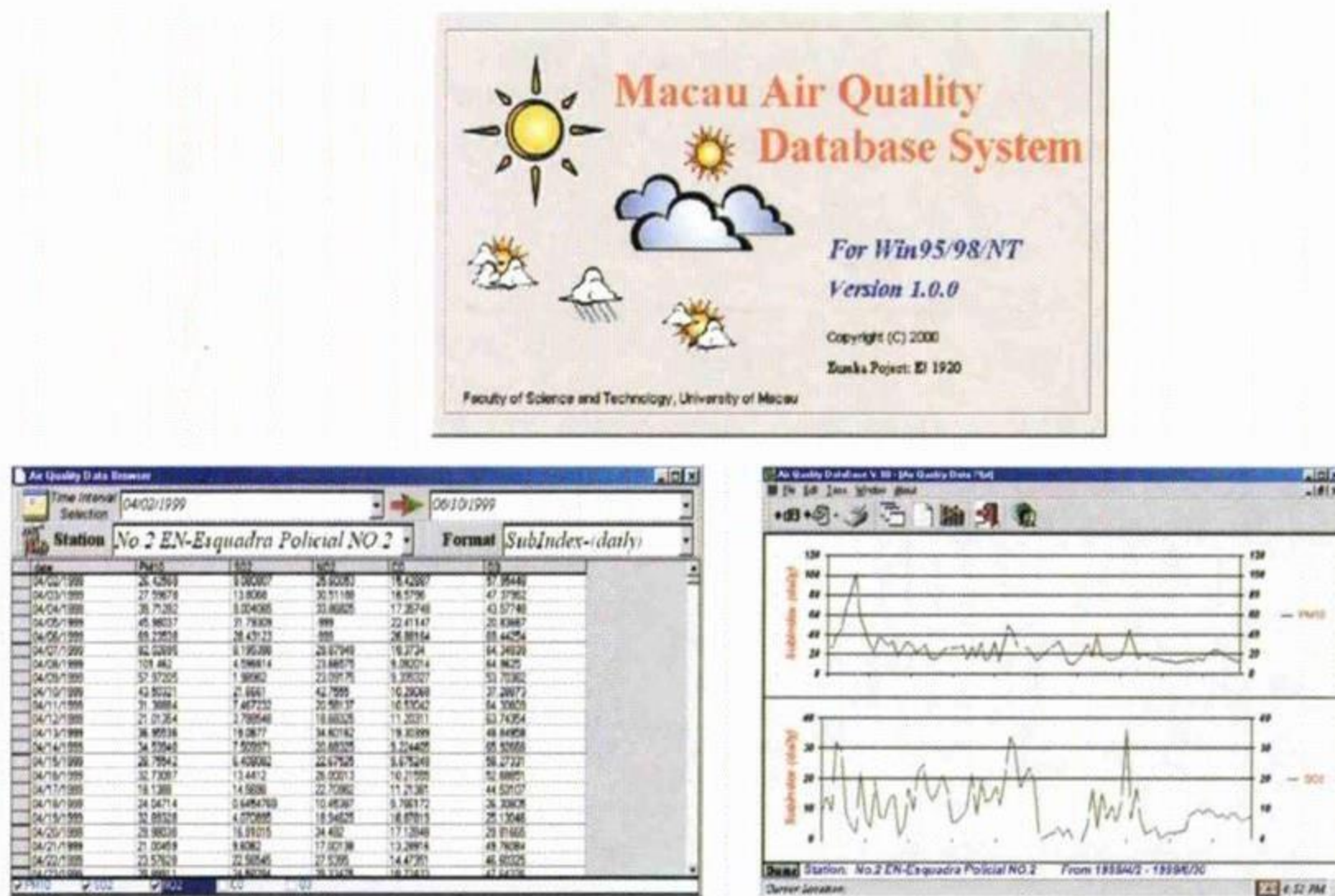


Figure 8. Views of the database management system interface.

Details of the complete system operation are reported by Mok et al.^[9,10,11] In short, the database allows the users to store the recorded pollutant concentrations and at the same time offers basic processing features such as data viewing, average calculation, episode identifying, AQI conversion and customized data output. The neural unit is developed for AQI forecasting. It uses a feed-forward neural network based algorithm trained by past AQI record. In other words, forecast of the future AQI is based on the knowledge of its history. The artificial neural network used in the forecasting system is the one described in Section 4 and tested in Section 6.1.

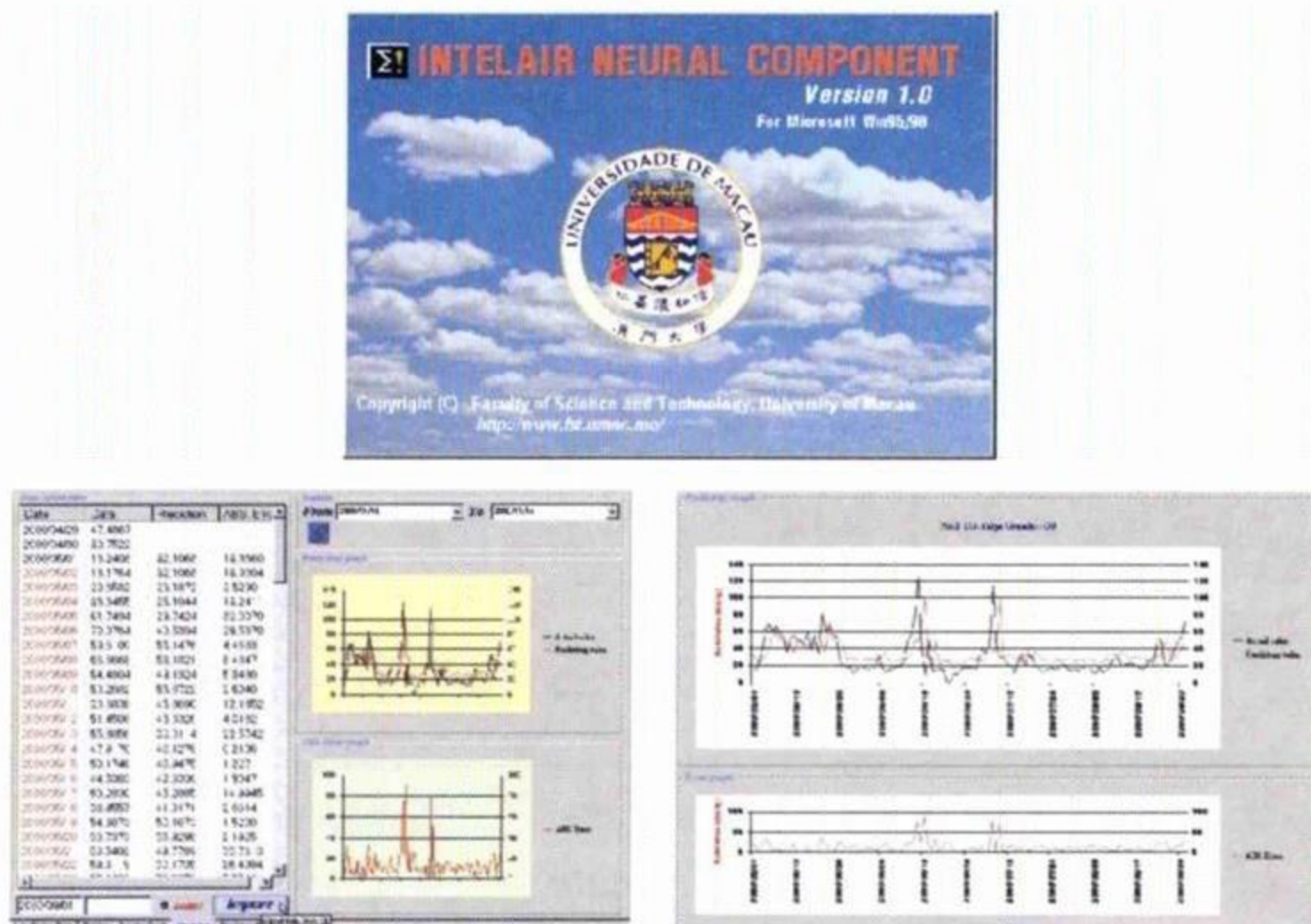


Figure 9. Views of INTELAIR Neural Component interface.

6.2.1. Application in Air Quality Index Prediction

To test the INTELAIR Neural Component on air quality forecasting ability, a time series of daily ozone sub-indices from the Taipa Grande Station in Macao was selected. The length of the data was from May 1, 1999 to August 31, 2000. For model training and performance evaluation, the data were divided into two segments. The data from 05/01/1999 to 04/30/2000 were utilized for model training and testing, while the data from 05/01/2000 to 08/31/2000 were used for model verification as shown in figure 10.

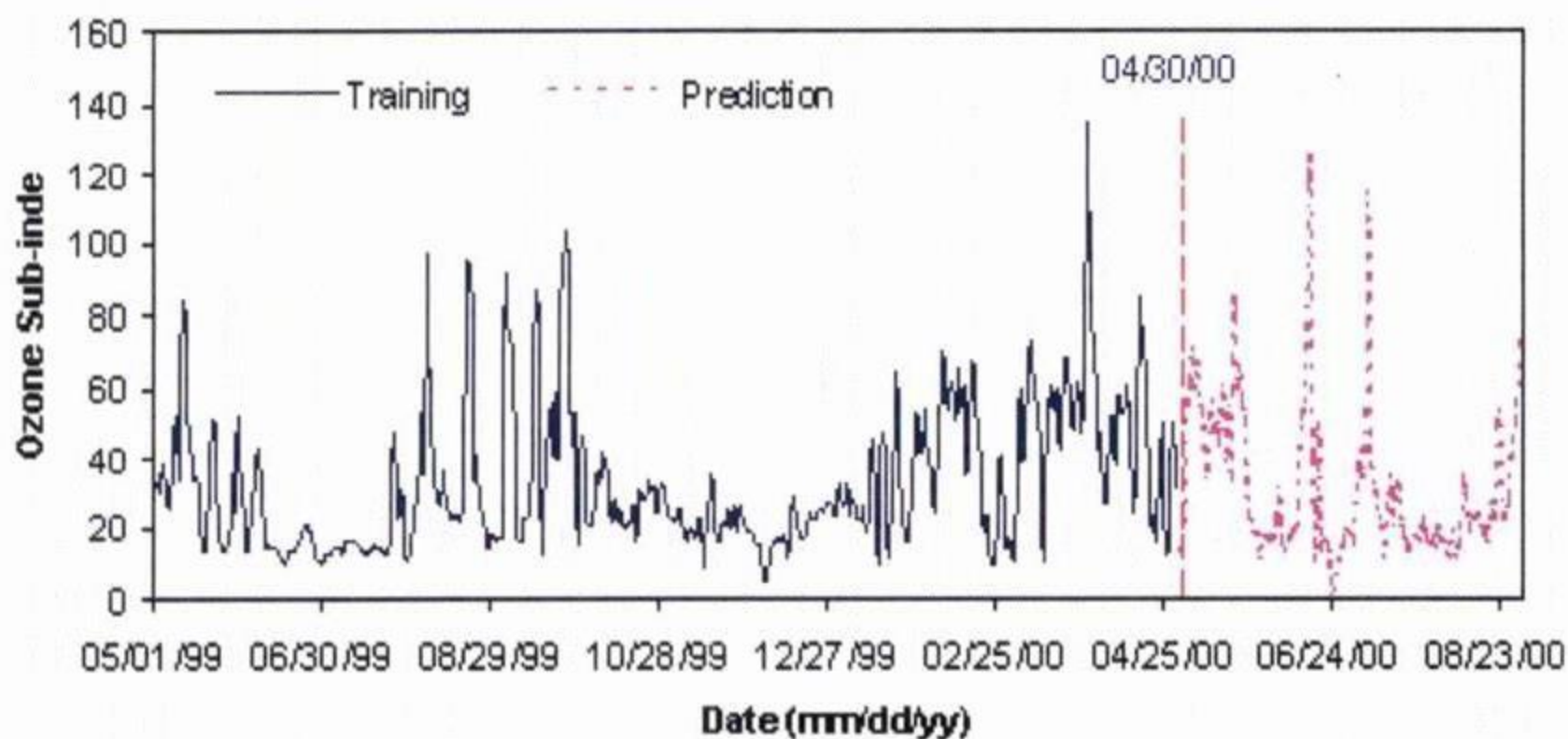


Figure 10. Variation of ozone sub-index from 05/01/99 to 08/31/00.

In order to select the most efficient structure, combinations of different input patterns and their corresponding numbers of neurons in the hidden layer were tried systematically. The training results of the model with this parameter setting are shown in figure 11. The model seemed to learn quite well by picking up most of the large peaks. The prediction of the model was then compared with the actual measurement as shown in figure 12. The network outputs match quite well with the actual values for relatively higher sub-indices. However, it was found that the predicted values lag behind the actual values. Since it was believed that the structure shown may not be the optimal one, improvement could still be made by performing extensive experimental trials on the network structures.

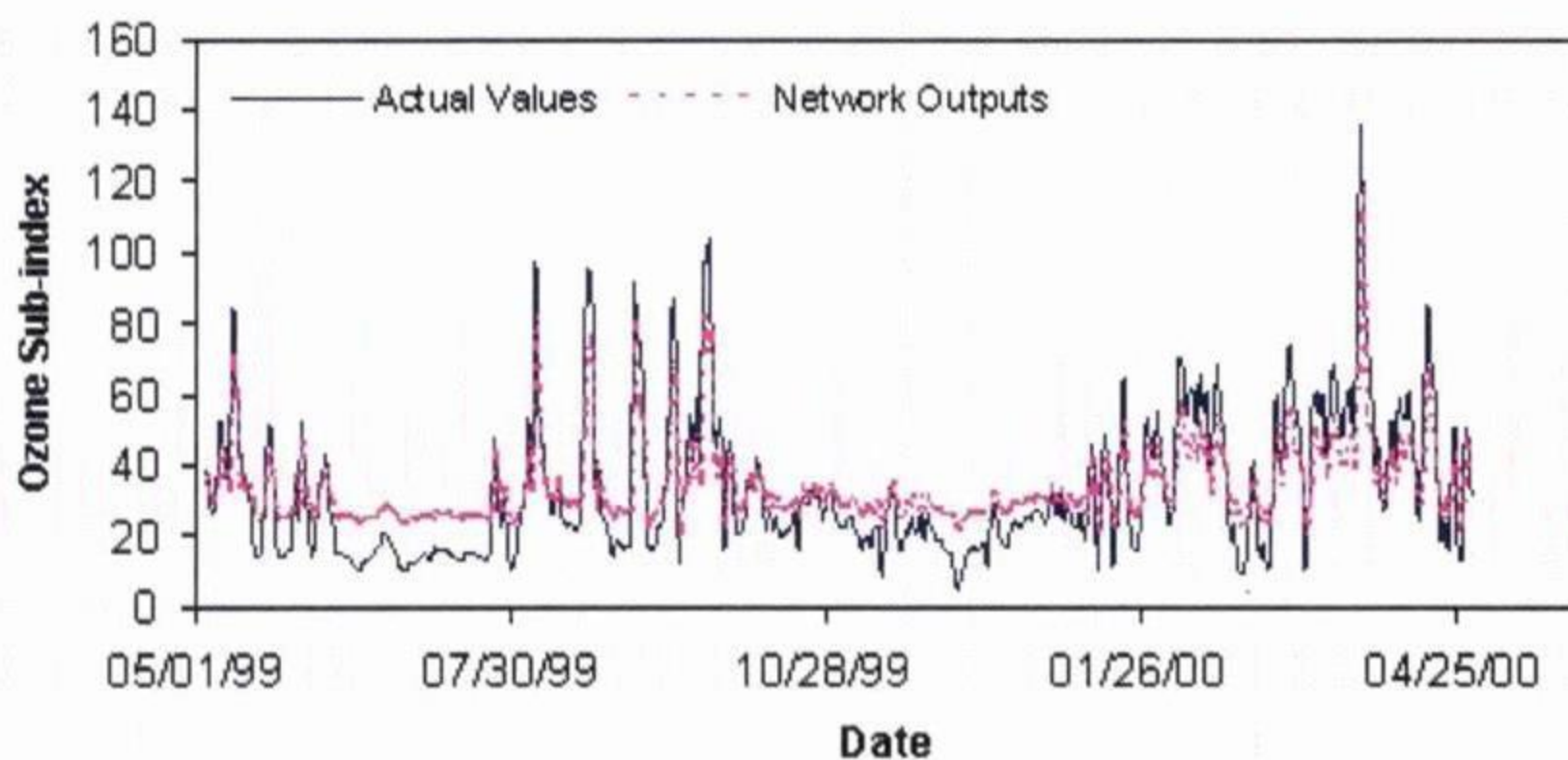


Figure 11. Performance of neural component on ozone sub-index training.

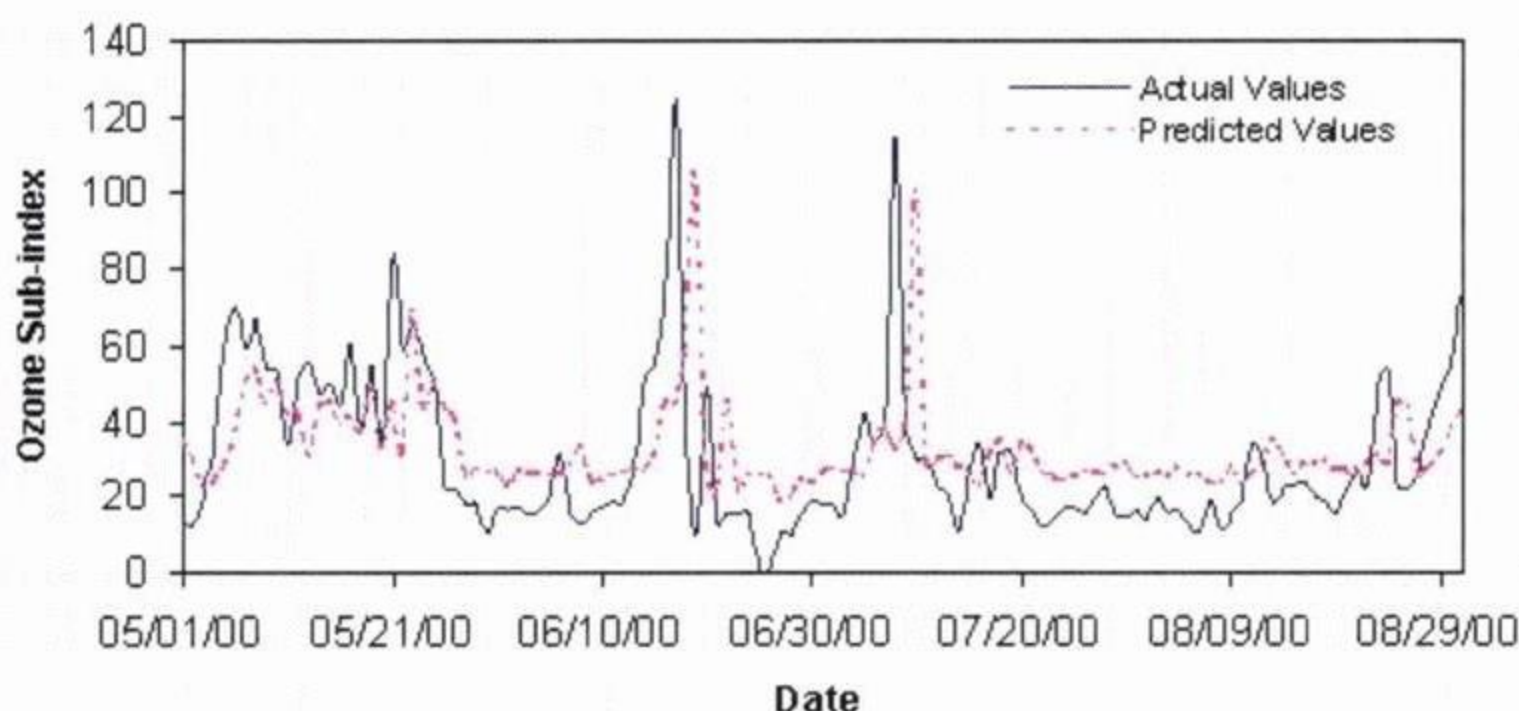


Figure 12. Performance of neural component on ozone sub-index prediction.

7. Adoption of System by Local Authority

The INTELAIR Neural Component was completed by the end of the year 2000 with great success. Since the system is very powerful, efficient and user-friendly, successful applications were found by the SMG during the period of February to May of 2001. The SMG then requested our software for their usage in releasing the daily forecasting AQI. An official transaction of the system from the University of Macau to the SMG was on June 8, 2001 (figure 13). On the same day, the system was also officially introduced to the general publics (figure 14).



Figure 13. Prof. Iu, Rector of University of Macau presenting the INTELAIR NEURAL COMPONENT to Mr. Fung, Director of SMG on June 8, 2001.



Figure 14. Prof. Mok presenting the INTELAIR report and introducing the UM developed neural component to the publics in June 8, 2001.

8. Concluding Remarks

Through studying the air quality situation in Macau, it was identified that the application of traditional air quality modeling techniques may not be suitable at the time. Not giving up on research, it was found through redefining the goals of research by looking into the actual needs of Macau that an air quality forecasting system was in demand. Successful research in application of artificial neural network on air quality prediction led to international recognition and a EUREKA project was resulted. The project finally led to the successful development of a user-friendly and efficient database and intelligent forecasting system for Macau air quality management, which is finally adopted for official usage by the local authority. The entire process involved both researches and development, which lasted for a period of approximately four years and the participation of personnel from different disciplines including civil engineering, mathematics and electrical and electronics engineering. The inter-discipline cooperation experience, and innovative ideas in application of artificial intelligence in air quality study made the road of the entire project both fun and challenging. It is well worth for sharing with the readers and hopefully these experiences would be helpful to those who may be going through similar endeavor in their research and development paths.

9. Acknowledgements

The development of the air quality forecasting system for Macau involved the cooperation and assistance of many personnel and units. Prof. S. C. Tam is the co-principle investigator of the research and development projects discussed in the present paper. Other personnel involved in the scientific and technical assistance are HOI Ka In, YAN Ping, LAM Leong Hong, LEI Soi In, IEONG Ka Kit, WONG Hou and CHAN Choi Iong. The studies were supported by the Research Committee of University of Macau under grant numbers RG002/97-98W/MKM/FST, RG002/97-98S/MKM/FST, and RG025/99-00S/MKM/EUREKA-INTELAIR. The Macau Meteorological and Geophysical Bureau is thanked for supplying the air quality data and the kind support by them in supplying detailed information on the Macau AQI system is highly appreciated.

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Apparent Molecular Weight Distribution of NOM in Macau Water and its Relation with the THM Formation

Wong Hou, Lou Wei Man, The Macau Water Supply Co. Ltd.

Li Shuang, Zhang Xiaojian, Tsinghua University

K. M. Mok, Wang Zhishi, University of Macau

(Faculty of Science and Technology, University of Macau)

(Laboratory and Research Center, Macau Water Supply Company)

Abstract: The performance of the Macau water treatment plants in the removing various apparent molecular weight (AMW) fractions of naturally occurring aquatic organic matter and humic substances is described. Ultrafiltration method is applied in this research for separation of the NOM in apparent molecular weight base. It is proved that Macau raw water has a very low TOC value and the apparent molecular weight lower than 3000 is the dominate fraction. As a general rule, THM reactivity ($\text{mg THM} / \text{mg C}$) increased with AMW.

Keywords: AMW, THM.

1. General Introduction

Surface water and, to a lesser extent, groundwaters contain naturally occurring dissolved organic matter (NOM), including humic substances. Thurman et al. indicated that the aquatic humic substances generally account for about 30% - 50% of the dissolved organic carbon in natural water sources. Aquatic humic substances are important from a water treatment perspective for several reasons, including their role as precursors in the formation of chlorination byproducts such as trihalomethanes (THMs) and other organo-Cl compounds. In addition, humic substances may be effective in the concentration and transport of inorganic and organic pollutants. Humic substances also complete with regulated chemicals for adsorption sites on activated carbon and can cause fouling of ion exchange and reverse osmosis membranes. Because of the problems associated with humic substances, their removal during water treatment must be optimized. Coagulation provides substantial removal of humic substances. The variability of THM precursor removal by water treatment plants can be due to both operation conditions as well as the specific characteristics of the humic material itself. Researchers (Oliver, B. et al. 1980) have shown that the THM yield or reactivity of humic substances from a given source as expressed in $\mu\text{g THM} / \text{mg C}$, varies as a function of molecular weight (AMW). Molecular weight for aquatic humic substances has been reported to range from 500 to 100000 Daltons. Thurman et al. (1982) suggested that this wide variation maybe due to the source of the humic substances, the analytical methodology used, and the aggregation of humic substances.

Since the city does not have large plenty of fresh water resource, source water (raw water) is taken from the Modaomen which is one of the eight widest estuaries of the Pearl River System. It is a main outlet of West River to the South China Sea. The runoff of West River accounts for 70% of the total runoff of Pearl River while that of Modaomen accounts for 28.67% of that of West River. The runoff of Modaomen is the biggest one in all the eight estuaries. The main water treatment plant located at Ilha Verde in Macau (maximum daily water supply 175000 m^3) applied the conventional water treatment technique, which includes pre-chlorination, coagulation, flocculation, sedimentation and filtration then final disinfection by chorine. Raw water is mainly supplied by the Mainland China. According to different water treatment technique of the plant, the efficiencies of each of it will be evaluated by comparing the AMW of the NOM before treatment and after the treatment, meanwhile, the raw water characteristics is analyzed. The water sample we used to perform the ultrafiltration UF

sample is the main water treatment plant in Ilha Verde, water samples during each treatment step was analyzed.

2. Related Work

2.1. Ultrafiltration

Ultrafiltration involves the separation of molecules from a liquid phase according to molecular size. Migration of molecules through membranes is generally accomplished by a combination of advective flow and molecular diffusion. The flux of a solute is

directly related to the area of membrane, concentration gradient, and molecular diffusion, but is inversely related to temperature. (Brock, T.D. 1983)

One main problem of ultrafiltration is called concentration polarization, the deposition of macromolecules on the membrane surface which results in a gel layer that becomes the dominant resistance to flow. This will appeared as the slow downed flux rate. Thus, an important requirement when using a membrane for AMW determinations is to ascertain when gel-layer diffusion, indicating that the membrane should be replaced or cleaned. As recommended by Macko et al and Brock, vigorous mixing of the feed solution is required in order to minimize the thickness of the membrane surface. Another suggestion is to keep the concentration of the solute as low as possible. Another problem arise is known as the Donnan effect, in which one of the solutes is unable to permeate the membrane. The phenomenon may lead to an unequal distribution of ions across the membrane, and, because electrical neutrality must be maintained, the membrane develops a potential that trends to impede the diffusion of any more ionic species that are similar in charge to that of the membrane. It is later suggested by Macke that the solution's pH and ionic strength should be held constant for uniform result.

The reason for selecting ultrafiltration is not only because of the easy solution of the induced technique problems and availability of equipment, but also due to another important requirement, the fractionated water sample according to different AMW after the ultrafiltration can be used to perform the THMFP test, this is main character of water sample of ultrafiltration which is totally cannot be satisfied by other techniques. The other methods can only provide the AMW distribution of analyzed water sample while cannot separate the different fraction of water sample accordingly. The advantage of ultrafiltration method attracts us to make this decision.

According to the literature, 75% of the THMs formed were derived from organics having a molecular weight of < 3000 Daltons. Moreover, 7% of the organics and 20% of the THMs were derived from compounds of <1000 Daltons molecular weight, in a related study, Veenstra and Schnoor (1980) found that the greatest THM yield per unit of organic carbon occurred in conjunction with molecules with an apparent molecular weight of <1000 Daltons. The molecular weight of aquatic humic substances generally ranges from approximately 500 – 10000 Daltons. The UF results for both raw and treated waters provided indicate relatively small differences between the THMFPs of the < 10000 Daltons fraction, the < 30000 Daltons fraction, and the overall humic content of the water (i. e., the 0.45 μ m filtrate). This result suggests the presence of little precursor material with an AMW of greater than 10000.

3. Experimental Method

3.1. Water Sample.

Four water samples was taken along the water treatment process, influent water (raw water and source water), coagulated water, filtrated water and treated water.

3.2. Molecular Weight Determination

The apparent Molecular weight of the NOM in the water is determined by the membrane ultrafiltration method. Before the ultrafiltration, raw water and the coagulated water is filtrated by the 0.45µm membrane (Millpore, Φ47 mm.) to remove the suspended solid. Apparent molecular weight distributions were obtained using a stirred cell (Amicon Inc, model 8400, effective volume 350ml) with membranes characterized by nominal AMW cutoffs of 1000, 3000, 10000, 100000 (YM1, YM3, YM10, YM100; Amicon Inc.). Sample aliquots were processed through the range of membranes in parallel, yielding a series of permeates, each containing all molecular weight fractions below the indicated

AMW cutoff, all permeates were characterized according to DOC, UV absorbance and THMFP.

3.3. Organic Carbon and THM Analysis.

Each step of the laboratory experiment is following the company SOP which is certificate by the CNACL 201-99 (equal to ISO/IEC). The water samples were sampled by dark glass bottles which were baked to the temperature of 500°C. The permeates test for the DOC (Shimadzu Int, model number TOC-5000A.) and UV254 (Shimadzu Int, model number UV-2401) after the filtration. During the THMFP experiment, the permeates were contained in a 50ml dark glass bottle and the initial chlorine concentration is about 20ppm. Incubated for 3 days under the temperature of 25°C in darkness then adding Na₂SO₃ to stop the reaction. The final water sample is being used for the testing of final THM. All water sample is analyzed in the company laboratory. The THM concentration is analyzed by the HPGC, which is the product of Perkin-Elmer.

4. Results and Discussion

Besides the raw water, the other water samples taken practiced prechlorination. A summary of chemical addition and operating conditions of water samples are presented in Table 1.

Table 1 Results of Ultrafiltration.

	Chlorine Dosage	UV254	DOC	SUVA	THMFP
	ppm	m ⁻¹	ppm	m ⁻¹ /mg	mol/L*100
Raw water (IR)	20	2.88	1.363	2.113	114.671
YM100	20	2.8	1.481	1.891	85.096
YM10	20	2.74	1.367	2.004	84.775
YM3	20	2.02	1.083	1.865	74.544
YM1	20	1.7	0.686	2.478	68.783
Coagulated Water (IM)	20	2.52	1.408	1.790	61.010
YM100	20	1.92	1.274	1.507	73.778
YM10	20	1.84	1.164	1.581	68.008
YM3	20	1.72	0.898	1.915	61.706
YM1	20	1.32	0.824	1.602	60.841
Filtrated Water (IF)	20	1.74	1.277	1.363	70.258
YM100	20	1.7	1.192	1.426	67.177
YM10	20	1.64	1.048	1.565	68.182
YM3	20	1.1	0.801	1.373	63.041
YM1	20	0.74	0.705	1.050	62.292
Treated Water (IT)	20	1.58	1.261	1.253	56.124
ITYM100	20	1.5	1.303	1.151	54.403
ITYM10	20	1.5	1.017	1.475	51.741
ITYM3	20	1.3	0.797	1.631	57.393
ITYM1	20	0.62	0.752	0.824	52.879

4.1. The apparent molecular weight (AMW) distribution of DOC, SUVA and THMFP along water treatment process.

The result of the apparent molecular weight distribution of the DOC, SUVA and THMFP variance along the water treatment process is presented in the Figure1, Figure 2 and Figure 3. DOC concentration of the Macau raw water is about 1.5~2 ppm normally, without much big variance along a year, the UV254 value is about 2.5~3.5 m^{-1} , accordingly, the SUVA value is always well below 3. According the statement of Edzald, this type of water has mainly the nonhumic material existed in the organic matter. The dissolved material is hydrophilic and comparatively less aromatic material existed in the small molecular weight organic materials. The coagulant has low effect on the remove of the DOC.

From Figure 1, the DOC remove efficiency is lesser than 20%. Especially for the DOC which molecular weight lowers than 1000 Daltons, the water treatment process hardly has any effects on that fraction of permeates. However, the organic matter which has the molecular weight lower than 3000 Daltons occupies about 80% of the total DOC concentration, almost 50% of the total DOC has the molecular weight lower than 1000. According to the literature, These observation obeys the opinion of Edzwald, for the nonhumic type water, low molecular weight organic dominate the DOC, and the conventional water treatment process has low effect on remove it.

The specific ultraviolet absorbance (SUVA) of a water which is defined as UV absorbance (measured in cm^{-1}) time 100 divided by the dissolved organic carbon concentration (mg/L), has been found to be a good surrogate for the water's humic content (Edzwald and Van Benschoten 1990). In Figure 2, high SUVA value is observed at the low molecular weight range of raw water (IR), this may imply that at a lower molecular weight range, there are more conjugated double bonds available to react with chlorine to form DBPs. From the line of treated water (IT), the SUVA value has much lowed, this may due to the chlorination effect. Referring to the AMW lower than 3000 Daltons, the SUVA has been decreased a lot after the treatment, this evidence indicate the most of the humic material existed in low molecular weight fraction of raw water has been much oxidized by the chlorination to form DBPs.

The THMFP is defined as the formation potential of trihalomethane, which is a well-known disinfection by-product because of its high molar mass fraction. In Macau this species is much concerned in some specific situation (i.e. salt water intrusion during dry season.). Figure 3 shows the distribution of the THMFP on the AMW base. There are three important observations. First, there is a very significant decrease of THM precursors appears between the raw water (IR) and the coagulated water (IM), this provided the effect of the prechlorination and coagulation on removing the organic matter of molecular weight higher than 100000 Daltons. Unfortunately, the precursor with the AMW lower than 100000 Daltons did not share this benefit, it is clear to see for the precursors with molecular weight lower than 3000 Daltons, the concentration has not large change despite of the second chlorination effect of the treated water (IT). Second observation is the dominate AMW fraction of the precursors in treated water is the

molecular weight lower than 1000 Daltons, it occupies about 95% of the total precursors existed in treated water. Third observation is almost all the THM precursors is concentrated on the molecular weight around 1000 Daltons, this fraction is also the part which is most difficult to be remove by conventional treatment plant, has the dominate effect on the THM formation.

4.2. THM reactivity along water treatment process.

The THM reactivates or yields (expressed in terms of mg THM/mg C) is shown in Figure 4. The average reactivity of the THM is around 1.2% ~ 0.8% for the raw water and 0.8% ~ 0.5% for the treated water. The reactivity of the THM is increasing along the

Decreasing of the AMW, both in raw water and treated water while the AMW lower than 1000 Daltons has almost (for the IT AMW<3000, the reactivity is also very high.) the highest

reactivity. Coagulation has proved its effect on reducing the THM reactivity, both for the AMW > 100000 and lower than 1000 Daltons, the reactivity of THMs decreased according to coagulation (IM). These two same decreasing trends are caused by different reasons. For the part AMW > 100000 Daltons, the decreasing of reactivity is induced by the removing of larger organic matter by coagulation and flocculation process. However the AMW < 1000 Daltons, the decreasing reactivity is induced by the first and second chlorination oxidized the most of the humic acid which concentrated in the low molecular weight fraction of organic matter.

Focusing on the treated waters, it can be seen that the aquatic organic matter remaining after treatment in each molecular weight range was generally less reactive in formation THMs in comparison to the untreated water. These results suggest that all of the various treatments preferentially remove the most reactive fraction of precursor present in each molecular weight range (Michael R. Collins, Gary L. Amy).

When comparing the figure of SUVA and THM formation reactivity (Figure 5), the trending of each corresponding curve is alike. Indicate the evidence that the SUVA is a good indicator for the trending of the reactivity of THMs in both raw and treated water.

5. Conclusion

Molecular weight for aquatic humic substances has been reported to range from 500 to 100000 Daltons from the literature. About 80% of the aquatic organic matter in Macau raw water has the AMW lower than 3000 Daltons, more advance, for the portion of AMW lower than 1000 Daltons it occupies about 50% of the total aquatic organic matter. The SUVA value of Macau water is always blow 3, from this two observation, we would conclude that Macau has a kind of low humic content source water and the aquatic organic matter existed in source water is dominated by low molecular weight. This factor has constrain the efficiency of coagulation so that is why coagulation has no significant efficiency on removing the organic matter in it (removing efficiency around 10%), for the organic matter of AMW lower than 1000 Daltons, the removing efficiency is almost 0. The humic material is concentrated on the fraction of AMW lower than 3000 Daltons (70%), the total removing efficiency is around 40%, both coagulation (12%) and filtration have effects on removing the humic materials. It is also expect that the first and second chlorination should also has efforts, however, there is no enough evidence to prove it. In Figure 3, the portion of AMW lower than 1000 Daltons dominate the THMFP generation (50%). Coagulation can remove a large portion of organic matter with AMW higher than 100000 Daltons. The reactivity of THM can be indicated by the factor of SUVA, it is clear to see that the SUVA value increasing along the decreasing of

The AMW of organic matter. After the treatment, both the reactivity and the SUVA value have been lowered. According to the statement of Michael R. Collins, all of the various

Treatments preferentially remove the most reactive fraction of precursor present in each molecular weight range. Despite of the fact that most reactive portion is existed in the organic matter with AMW lower than 1000, the reactivity of the each fraction is decreased along the treatment.

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Figures

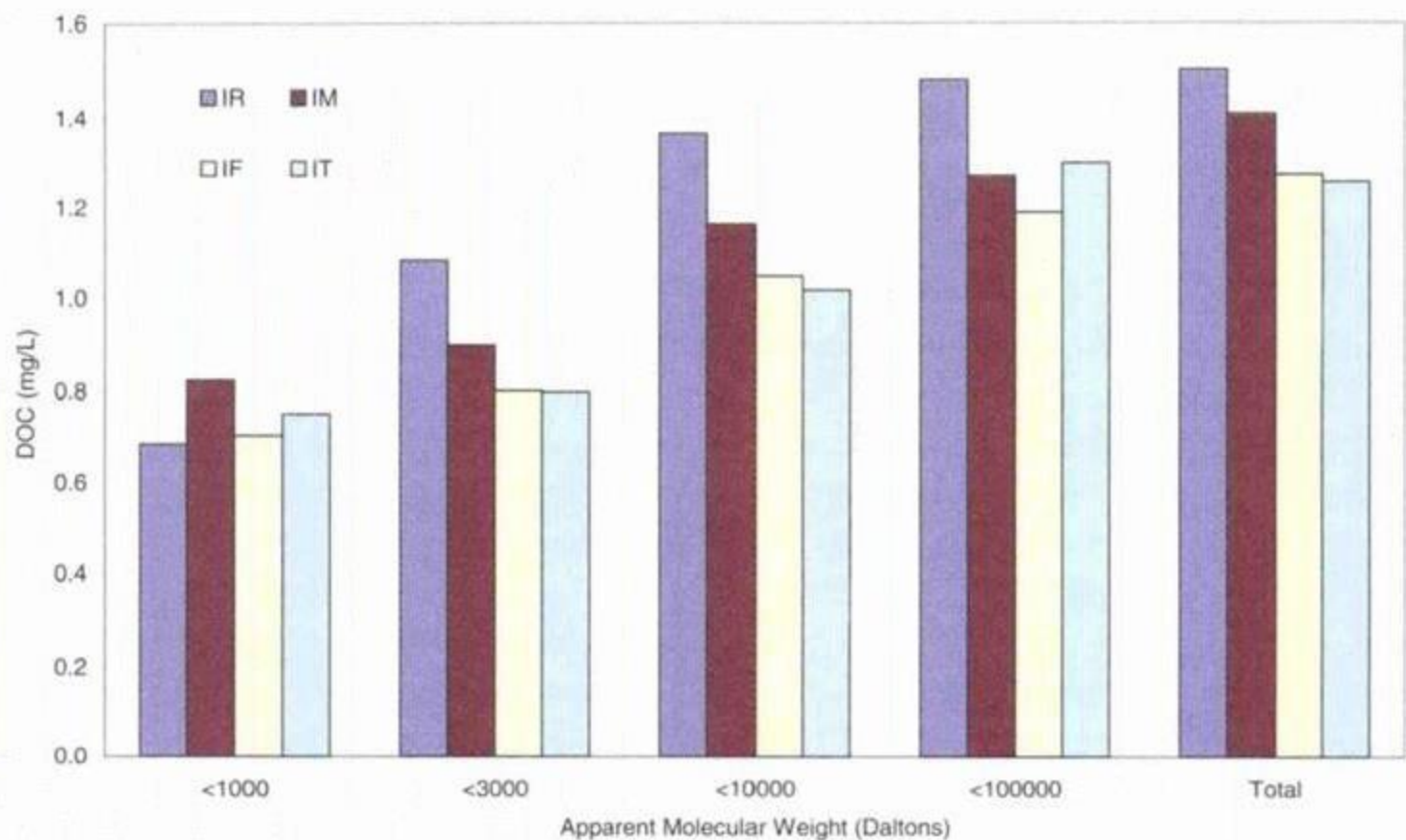


Figure 1. DOC variance along water treatment process.

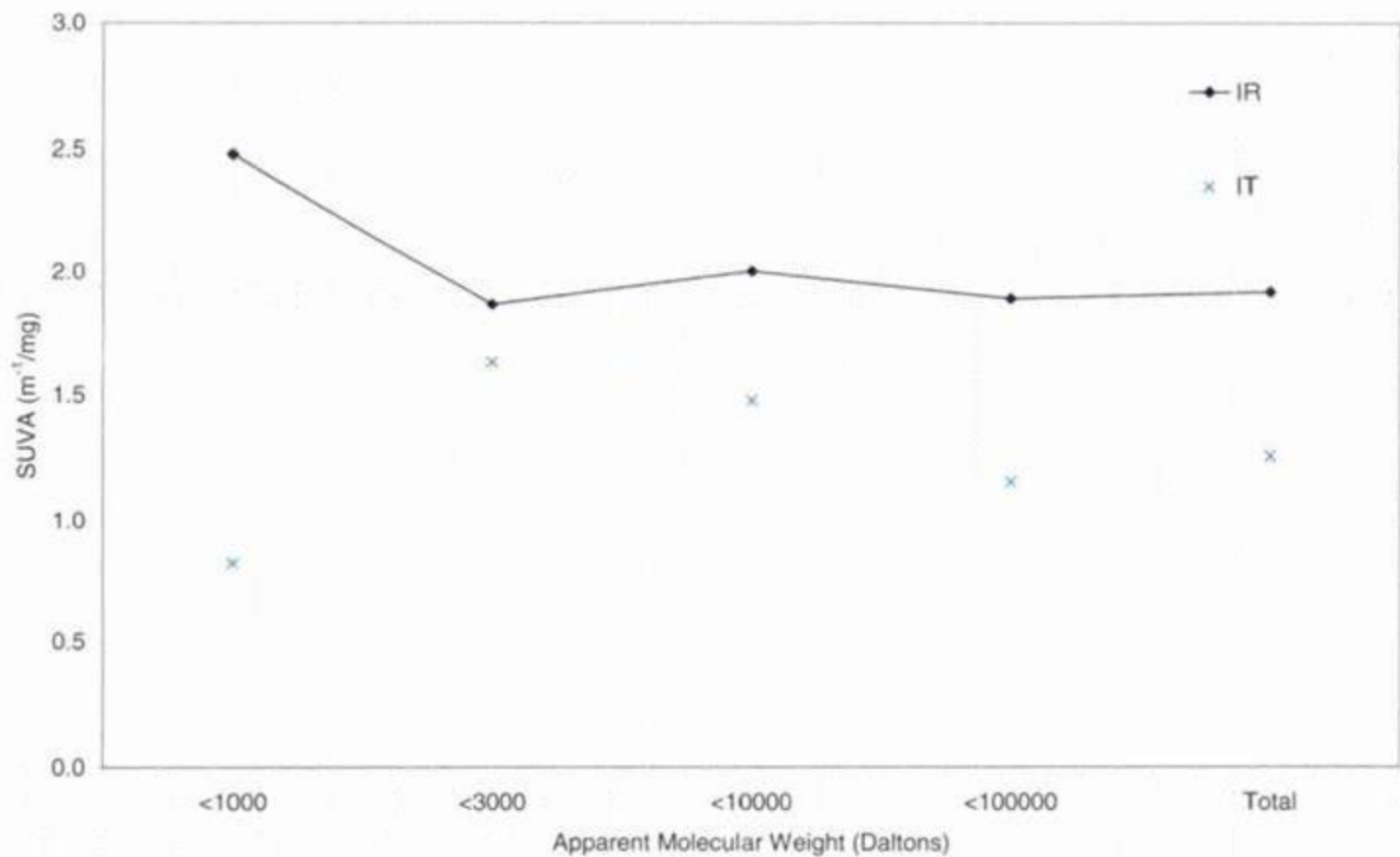


Figure 2 SUVA comparison of the influent water and treated water along treatment process.

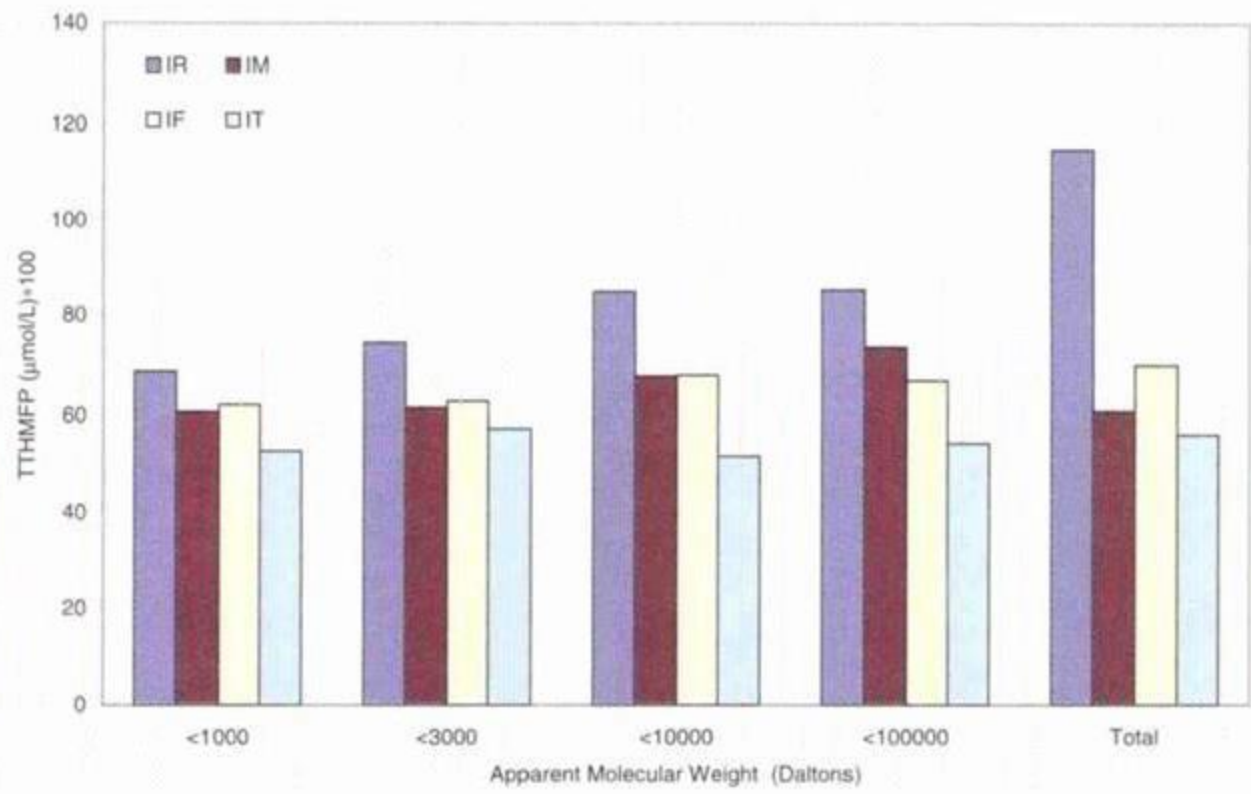


Figure 3 The THMFP variance along water treatment process.

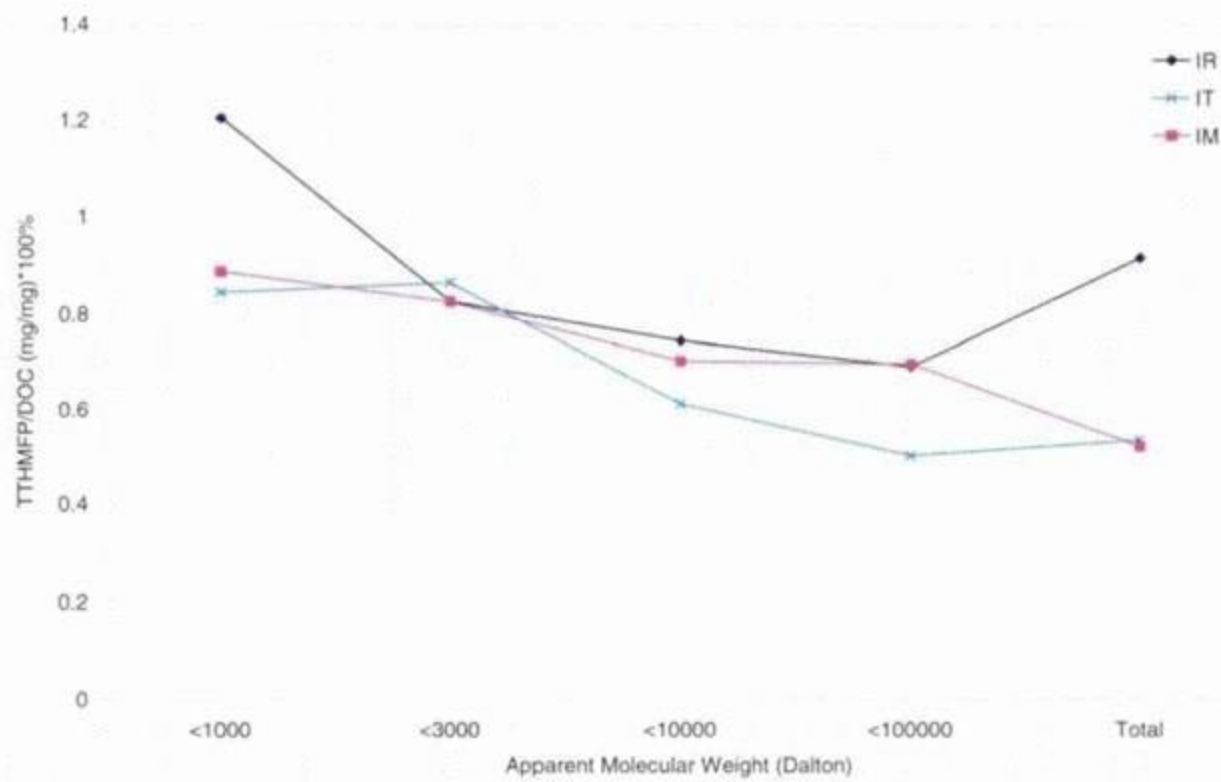


Figure 4 THM reactivity along water treatment process.

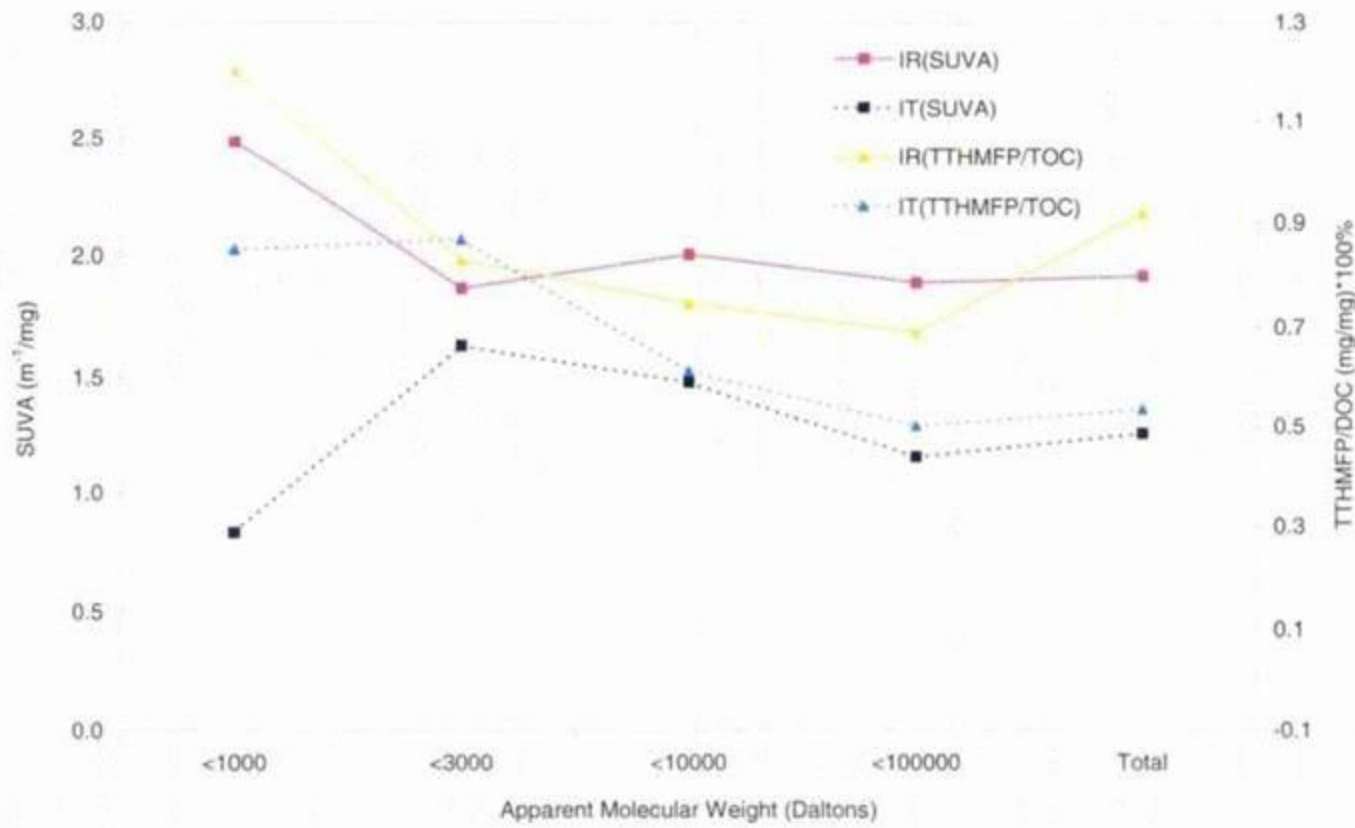


Figure 5 The comparison of SUVA and THM reactivity.

澳门交通环境颗粒物物理化学特征分析

郝吉明, 吴烨, 傅立新, 胡京南, 王丽涛

(清华大学环境科学与工程系, 北京 100084)

王志石, 邓宇华

(澳门大学科技学院, 澳门)

摘要: 通过现场监测和采样, 对澳门交通环境颗粒物基本的物理和化学特征进行了分析。采用 DustTrak 和 TEOM1400a 颗粒物监测仪对道路边颗粒物浓度的垂直分布、水平分布和粒径分布的监测结果表明: 1) PM_{10} 、 $PM_{2.5}$ 和 PM_1 的浓度均表现出了明显的随高度增加而递减的规律, 在 79m 处, PM_{10} 、 $PM_{2.5}$ 和 PM_1 的浓度分别衰减为地面最大浓度的 60%、62% 和 80%; 2) 道路边颗粒物水平方向上的衰减趋势并不明显, 在距道路边 0-228m 范围内, PM_{10} 、 $PM_{2.5}$ 和 PM_1 的最大浓度衰减幅度仅分别为 7%、9% 和 10%; 3) 道路边 $PM_{2.5}$ 和 PM_1 分别贡献了 PM_{10} 总浓度的 66-67% 和 51-60%, 表明澳门道路边细粒子和亚微米级粒子占据了 PM_{10} 的大部分份额。采用 TEOM1400a 和 ACCU 颗粒物系统对澳门道路边 PM_{10} 和 $PM_{2.5}$ 进行了采样, 其主要化学成分分析结果表明: 1) 有机成分是道路边颗粒物中最重要的成分, 其比例超过 30%; 2) SO_4^{2-} 等二次粒子、EC、地壳元素和海盐成分也是颗粒物的重要组成部分, 分别占 5-20% 不等的份额; 3) 在城市居住区和背景点 EC 浓度很低, 但在道路边, 特别是柴油车流量较大的交通环境, EC 比例很高, 这表明 EC 是反映机动车尤其是柴油车排放的标志性元素; 4) 初步分析结果显示道路边的 $PM_{10}/PM_{2.5}$ 来源比较复杂, 机动车排放、道路尘和餐馆油烟排放可能是重要的污染源, 此外二次粒子和背景浓度等较大尺度的区域性外来源也是很重要的影响因素。

关键词: 颗粒物, 垂直分布, 水平分布, 粒径分布, 化学组成, 车流

90 年代以来, 随着澳门人口的增长和经济的发展, 城市机动车保有量以年平均约 9.8% 的速度快速增长, 截至 1999 年底, 澳门的机动车总保有量已经达到 11.3 万辆, 成为世界上机动车密度最高的地区之一。由于澳门地区经济以商业和旅游业为主, 无重工业; 民用能源以石油气为主。因此, 机动车排放已成为澳门地区最主要的大气污染源, 由此造成的环境污染问题已日益突出。

与气态污染物相比, 交通环境颗粒物来源更为复杂。城市交通环境颗粒物的排放主要包括尾气管排放、道路二次扬尘、轮胎磨损、刹车磨损等等^[1], 特别是尾气管排放和道路扬尘的协同作用, 使得城市交通密集区域不论是粗颗粒的浓度或是细粒子的浓度都显著高于城市区域的平均浓度^[2,3]。

大气污染物浓度的空间分布特征是研究污染物传输、迁移和扩散规律的基本内容。道路边颗粒物浓度由于受到复合排放源、粒子特性、气象条件等多种因素的影响, 其空间分布规律较一般气态污染物 (CO 、 NO_x 等) 更为复杂。此外, 交通环境颗粒物排放源不仅来源复杂, 而且不同源排放颗粒物的粒径分布和化学成分也存在很大差异。道路扬尘通常被认为是粗粒子 ($2.5\sim 10\mu m$) 的最重要贡献源, 其标志性化学组分包括 Ca、Si、Al、Fe、Mn 等元素; 而机动车排放则是细粒子 ($PM_{2.5}$), 特别是亚微米级粒子 (PM_1) 的重要来源, 其主要成分包括 OC、EC 和 Pb 等^[4,5]。

本研究通过对澳门典型道路的颗粒物浓度空间分布监测, 获得不同粒径粒子 ($PM_{10}/PM_{2.5}/PM_1$) 垂直分布和水平分布的浓度廓线; 通过对道路边颗粒物粒径分布的监测, 获得了 $PM_{2.5}$ 和 PM_1 在 PM_{10} 中的比例。此外, 通过对道路边颗粒物采样的化学分析, 获得了 $PM_{2.5}$ 和 PM_{10} 基本的化学成分谱。通过上述的研究, 可进一步定性分析各种排放源对道路颗粒

物浓度的贡献水平。

1. 道路边颗粒物浓度空间分布和粒径分布规律研究

1.1. 监测方法和程序

本研究选取澳门的 3 条典型道路，分别进行了颗粒物浓度的空间分布和粒径分布监测，监测地点和监测内容如下：1) 高士德大马路：PM₁₀/PM_{2.5}/PM₁ 的垂直分布；2) 友谊大马路：PM₁₀/PM_{2.5}/PM₁ 的水平分布；3) 沙梨头海边街：PM₁₀/PM_{2.5}/PM₁ 的单点监测（粒径分布）；4) 友谊大马路：PM₁₀/PM_{2.5}/PM₁ 的单点监测（粒径分布）。

1.1.1. 监测仪器

1) DustTrak

DustTrak8520 是美国 TSI 公司制造的便携式颗粒物实时监测仪。该仪器利用激光光散射技术（散射光强与粒子体积浓度成比例）测定环境中颗粒物的实时浓度。DustTrak 配有 10μm、2.5μm 和 1μm 三种切割粒径的采样头，采样流量为 1.7L/min。由于 DustTrak 采用亚里桑那标准测试尘（Arizona Test Dust）来标定散射光强与粒子质量浓度之间的关系，因此，当监测环境颗粒物的特性与测试尘差别较大时，其读数并不能反映颗粒物的真实质量浓度。在实际使用过程中，需要对该仪器读数进行再标定。本研究中，DustTrak 每分钟计数，逐时平均。

2) TEOM

TEOM1400a 是美国 Rupprecht & Patashnik 公司制造的颗粒物实时监测仪。该仪器已通过美国环保局认证（EQPM-1090-079），并已成为联邦等效测试方法（Federal Equivalent Method, FEM）^[4]。TEOM1400a 采用振荡微天平技术（Tapered Element Oscillating Microbalance, TEOM）测定环境中颗粒物的真实质量浓度。该仪器配有 10μm 和 2.5μm 两种切割粒径的采样头，采样流量为 16.7L/min。本研究中，TEOM 读数取逐时平均。在分析交通环境颗粒物的粒径分布时，采用 TEOM1400a 对 DustTrak8520 进行再标定。

1.1.2. 监测程序

道路边颗粒物浓度垂直分布规律的监测地点选择在澳门高士德大马路和俾利喇街的交界处。按照距离地面高度的不同（2、8、19、30、59 和 79m）共布设了 6 个监测点，每个测点放置一台 DustTrak 监测仪。采样管从窗口垂直伸出至少 0.5 m。垂直分布监测点布设的示意图见图 1。考虑到绝大部分的车流量集中在白天，因此将监测时间设定为 8:00am-20:00pm，持续三天（2001 年 12 月 4 日-6 日）。其中 12 月 4 日监测 PM₁₀，12 月 5 日监测 PM_{2.5}，12 月 6 日监测 PM₁。

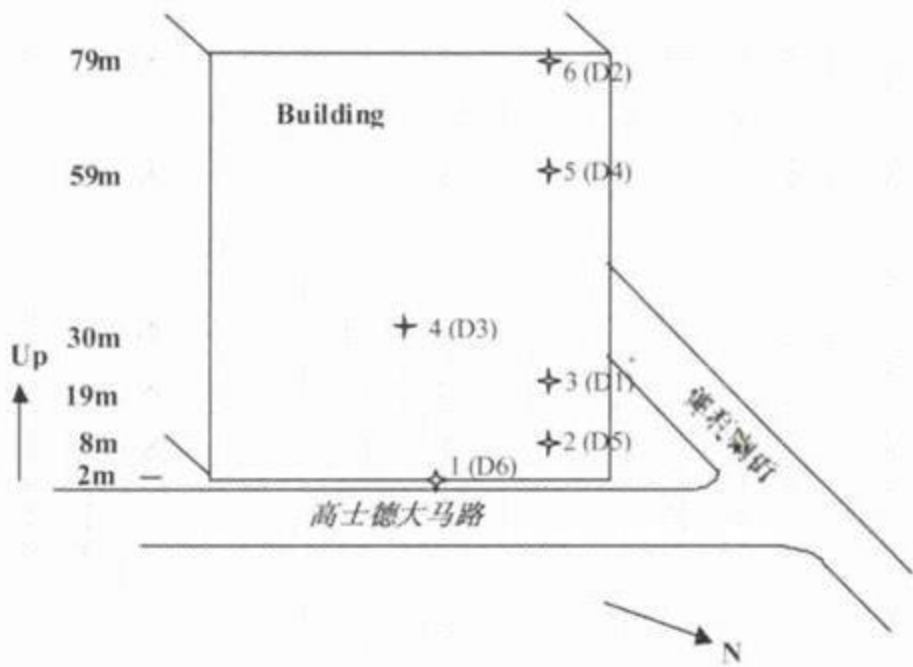


图 1 道路边颗粒物浓度垂直分布监测地点示意图

道路边颗粒物浓度水平分布规律的监测地点位于澳门友谊大马路。按照距离道路边水平距离的不同(2、42、72、120、170 和 228m) 共布设了 6 个监测点, 每个测点放置一台 DustTrak 监测仪。考虑到水平方向污染物浓度衰减最强烈的往往出现在距道路边 100~150m 范围以内^[6], 因此, 本研究在 150m 范围内共设置了 4 个监测点。仪器采样管距离地面的垂直距离约为 1.5~2.0m。水平分布监测点布设的示意图见图 2。基于类似的考虑, 将监测时间设定为 8:00am-20:00pm, 持续三天(2001 年 12 月 10 日-12 日)。其中 12 月 11 日监测 PM₁₀, 12 月 12 日监测 PM_{2.5}, 12 月 10 日监测 PM₁。

道路边颗粒物浓度的单点监测的地点共选择了 2 条典型道路: 1) 友谊大马路; 2) 沙梨头海边街。在每条道路的两边各布设两个监测点(距道路边 2~3m), 每个测点各放置三台 DustTrak 监测仪, 分别配备 10μm、2.5μm 和 1μm 的采样头; 同时在道路的南边放置 TEOM1400a 进行同步监测。DustTrak 和 TEOM1400a 采样管距离地面的垂直距离分别约为 2.5m 和 3.0m。友谊大马路监测时间为 2001 年 12 月 3 日 8:00am-20:00pm; 沙梨头海边街的监测时间为 2001 年 12 月 7 日 8:00am-20:00pm。道路边单点监测的示意图见图 3。

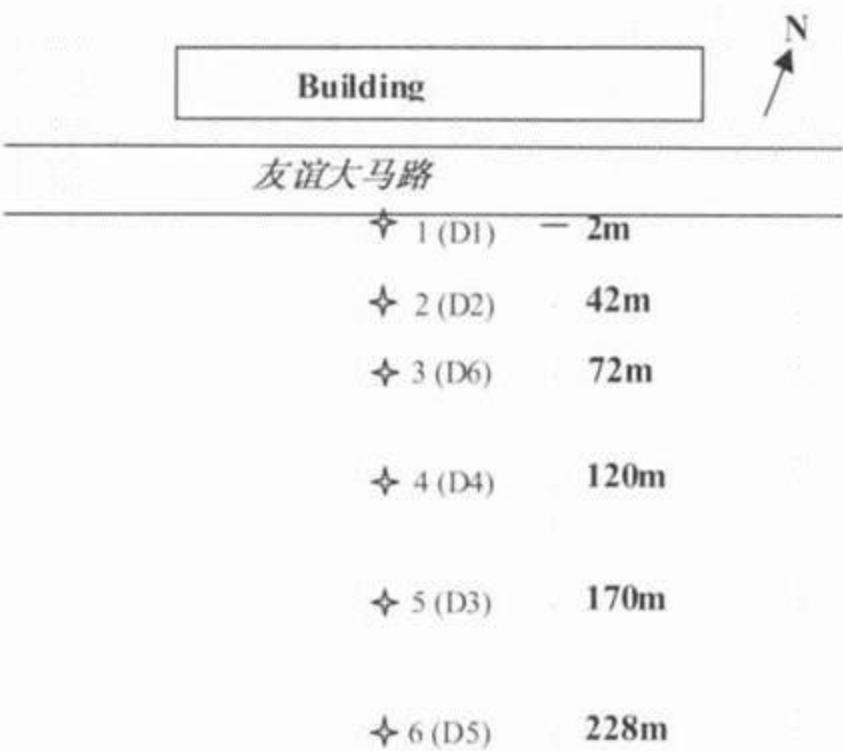


图 2 道路边颗粒物浓度水平分布监测地点示意图

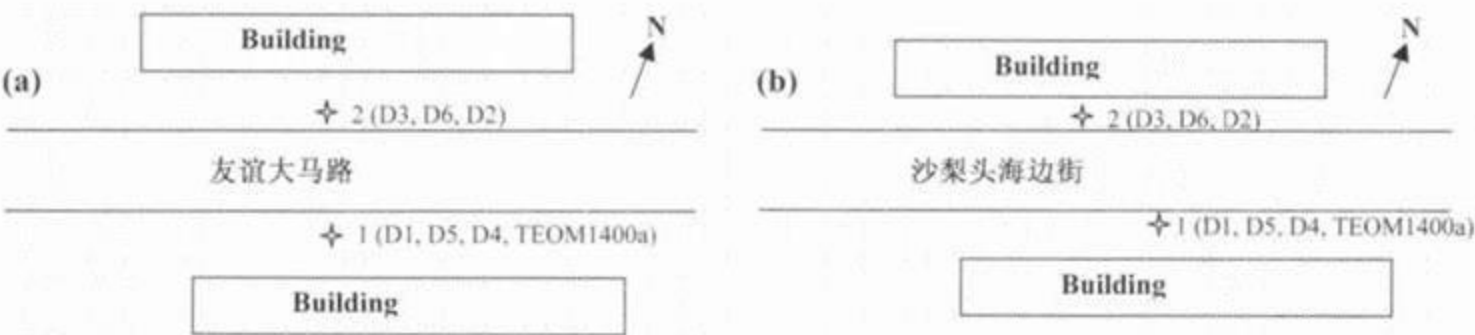


图 3 道路边颗粒物浓度单点监测示意图

在对颗粒物浓度分布进行监测的同时, 进行气象参数和车流状况的同步监测。在澳门, 选取大潭山的同步气象监测资料。监测的气象参数包括: 风向、风速、温度、相对湿度、气压和降雨量。车流统计将车辆分为轿车、出租车、摩托车、轻型车、重型车和公共汽车六类, 分别获得了各典型道路分车型和总车流量的逐时变化资料。

1. 1. 3. 质量保证体系

为了确保测试数据的准确可靠, 本研究在每次监测前对所有 DustTrak 监测仪进行流量校正、读数清零、清除切割头上的颗粒物并涂抹硅油; 对 TEOM1400a 进行流量校正, 并定期清除 PM_{2.5} 切割头中的颗粒物。

由于切割头和内部光学系统的微小差异, 导致不同的 DustTrak 读数存在系统偏差。因此, 本研究对仪器进行了平行监测, 分析结果见表 1。结果表明, 各仪器时均浓度之间的相关性非

常高，相关性系数 R^2 达到了 0.95~1.00。不同仪器的读数有所差别，其中 D2 和 D4 的 PM_{10} 时均浓度差别 10.4%。因此，为了消除不同仪器读数之间的这种系统偏差，以标号为 D2 的 DustTrak 监测仪的读数为基准，其余 5 台 DustTrak 监测仪的读数将按照表 1 中的 Ratio 值分别进行折算。

表 1 DustTrak 颗粒物监测仪的平行监测结果

DustTrak 编号	PM_{10}			$PM_{2.5}$			PM_1		
	Ratio ^a	R^2 ^b	n ^c	Ratio	R^2	n	Ratio	R^2	n
D1	0.945	0.992	14	0.953	0.997	8	0.972	0.999	5
D3	1.030	0.960	6	0.977	0.989	8	0.972	1.000	8
D4	0.928	0.946	6	0.918	0.994	8	0.896	1.000	8
D5	0.968	0.999	8	0.924	0.989	8	0.943	1.000	6
D6	0.984	0.950	6	0.973	0.996	8	0.994	1.000	8

^aRatio=其它 DustTrak 监测仪的浓度读数与编号为 D2 的 DustTrak 的浓度读数的比值
^b R^2 =相关性系数
^cn=逐时平均的样本数

1.2. 道路边颗粒物浓度的垂直分布规律

道路边颗粒物浓度的垂直分布变化见图 4。结果表明， PM_{10} 、 $PM_{2.5}$ 和 PM_1 的浓度均表现出了明显的随高度增加而递减的规律，最大浓度无一例外的出现在地面监测点上（点 1）。

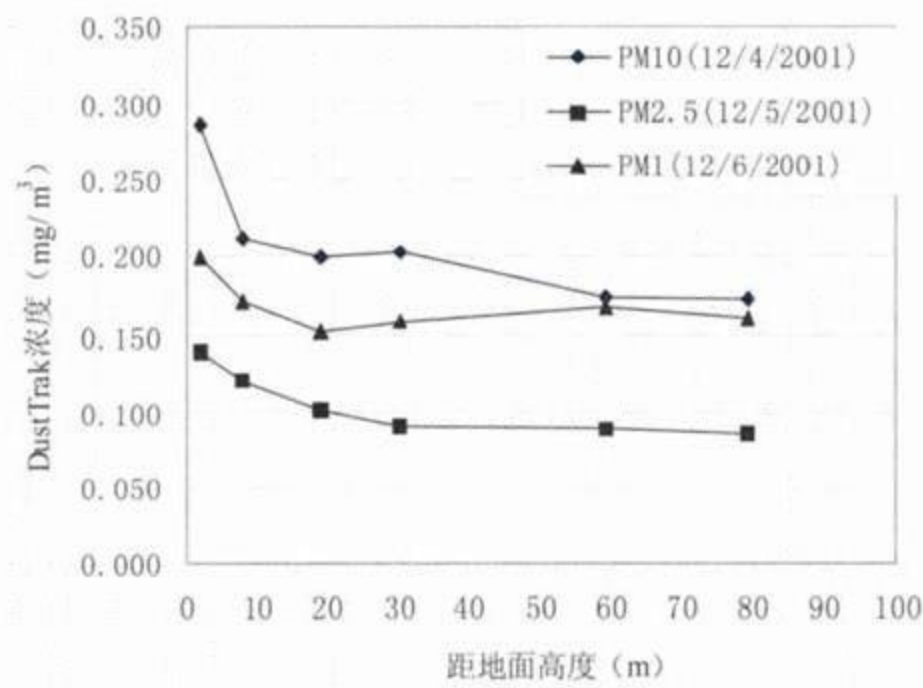


图 4 道路边颗粒物浓度（监测期间平均值）的垂直分布

在垂直分布上， PM_{10} 浓度表现出了很强的衰减趋势，特别是在近地面区域。在从距地面 2m 到 8m 高度这一范围内， PM_{10} 平均浓度衰减了 26%。但是，随着高度的继续增加， PM_{10} 浓度衰减幅度大幅度的趋缓。在 79m 处（点 6）， PM_{10} 平均浓度是地面浓度（点 1）的 60%。也就是说，从距地面 8m 到 79m 高度这一范围内， PM_{10} 平均浓度总共仅衰减了 14%。

在垂直分布上， $PM_{2.5}$ 浓度也表现出了很强的衰减趋势，但是其变化规律与 PM_{10} 有所不同。在从距地面 2m 到 8m 高度这一范围内， $PM_{2.5}$ 平均浓度仅衰减了 13%，不如 PM_{10} 明显。但是，随着高度的继续增加， $PM_{2.5}$ 浓度依然保持较大的衰减幅度。在 19m 和 30m 高度处，浓度的衰减幅度分别为 27%和 34%。30m 以上的区域，浓度的变化已不明显。在 79m 处， $PM_{2.5}$ 平均浓度是地面浓度的 62%。

PM₁ 浓度在垂直分布上的衰减趋势则不如 PM₁₀ 和 PM_{2.5} 显著。在从距地面 2m 到 8m 高度这一范围内, PM_{2.5} 平均浓度衰减了 14%, 与 PM_{2.5} 类似。在整个的监测高度内, 浓度衰减幅度最大的点出现在 19m 处, 为 23%。随着高度的增加, PM₁ 浓度反而有所上升。在 79m 处, PM₁ 平均浓度是地面最大浓度的 80%。

道路边颗粒物浓度垂直分布的这种递减规律表明, 无论是粗粒子还是细粒子, 都受到了地面交通源的显著影响。当然, 不同的排放源对不同粒径粒子的影响程度是有差异的。道路扬尘可能是影响道路边粗粒子浓度的最重要贡献源; 而对于细粒子, 特别是亚微米级粒子, 机动车排放也是重要的来源。

对于 10μm 以下的粒子, 其重力沉降速度可以用 Stokes 公式来表述^[7]:

$$u_s = \frac{d_p^2 \rho_p}{18\mu} gC \quad (1)$$

式中, u_s 为重力沉降速度, d_p 为颗粒物粒径, ρ_p 为颗粒物密度, μ 为粘滞系数, C 为坎宁汉修正系数 (1μm 以上的粒子可以不予考虑)。对于 1~10μm 的颗粒物, 其重力沉降速度与粒径的平方成正比, 而对于要考虑坎宁汉修正的粒径小于 1μm 的粒子, 其重力沉降速度与粒径依然成正比。因此, 粗粒子的重力沉降速度要比细粒子大得多, 这也是导致 PM₁₀ 浓度在近地面区域衰减趋势远大于 PM_{2.5} 和 PM₁ 的重要因素。

1.3. 道路边颗粒物浓度的水平分布规律

道路边颗粒物浓度的水平分布变化见图 5。结果表明, 虽然 PM₁₀、PM_{2.5} 和 PM₁ 的最大浓度值都出现在距道路最近的监测点上 (点 1), 但是其浓度随着距道路水平距离的增加而递减的趋势并不明显。

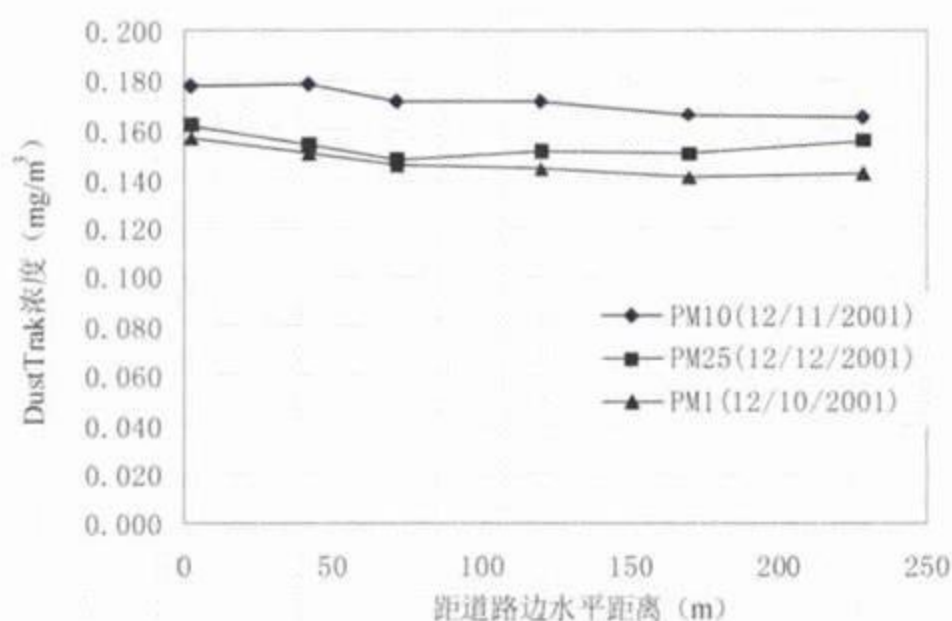


图 5 道路边颗粒物浓度 (监测期间平均值) 的水平分布

在水平分布上, PM₁₀ 浓度的衰减趋势并不明显。在整个水平监测范围内 (2~228m), PM₁₀ 平均浓度最大衰减幅度仅 7%, 出现在距道路边 228m 处 (点 6)。并且, 有多个监测点之间的浓度 (例如点 1 和点 2, 点 3 和点 4, 点 5 和点 6) 在统计上无显著性差异。

在水平分布上, PM_{2.5} 浓度的衰减趋势也不明显。在整个水平监测范围内, PM_{2.5} 平均浓度最大衰减幅度仅 9%, 出现在距道路边 72m 处 (点 3); 且随着道路边水平距离的进一步增加 (72~228m), 浓度反而有所增加。在监测期间, 风向发生了较大的改变, 由早上的从友谊大马路吹向监测点转变为下午的从监测点吹向友谊大马路。显著性差异的统计结果表明, 下午各监测点之间的浓度, 除了道路边的点 1 之外, 并无差异。这是由于风向改变之后, 各监测点处于上风向, 除了点 1 位于道路边, 受到道路的影响较大外, 其余各点的浓度仅反映了城区背景点的水平, 因此在数值上并无统计差异。然而, 下午监测期间点 1 PM_{2.5} 的平均浓度也仅比

其余各点的平均浓度高出 12% 左右。

由于 12 月 10 日清晨有降雨过程，因此 PM_{10} 的监测从午后路面干燥之后开始。结果表明， PM_{10} 浓度的水平递减趋势也不明显。在整个水平监测范围内， PM_{10} 平均浓度最大衰减幅度也只有 10%，出现在距道路边 170m 处（点 5）。

由于采样高度设置比较低（1.5~2.0m），因此水平分布的监测容易受到各监测点周边局地源的干扰，这可能是造成水平分布递减规律不明显的因素之一。此外，由于友谊马路车流量并不高，因此该道路上机动车排放和二次扬尘产生的颗粒物浓度在该区域总浓度（包括背景浓度和其它源的贡献）中贡献并不突出也可能导致水平分布不明显。国外的相关研究也表明，道路边水平方向颗粒物浓度的衰减幅度较小，TSP、 PM_{10} 或 $PM_{2.5}$ 的浓度在距道路边 0~400m 范围内一般减少 0~25% 左右^[5,8,9]，道路边最大浓度与城区背景浓度相比一般也仅高出 30% 左右^[2]。

1.4. 道路边颗粒物浓度的粒径分布规律

由于 DustTrak 采用亚里桑那标准测试尘来标定散射光强与粒子质量浓度之间的关系，因此，当监测环境颗粒物的特性与测试尘差别较大时，其读数并不能反映颗粒物的真实质量浓度。本研究在分析交通环境颗粒物的粒径分布时，采用 TEOM1400a 对 DustTrak8520 进行再标定。图 6 给出了澳门道路边 DustTrak 和 TEOM 监测仪逐时浓度的比对结果。

DustTrak 和 TEOM 监测仪的逐时浓度有很好的相关性。对于 PM_{10} ，DustTrak 的逐时浓度是 TEOM 的 2.0 倍，两者浓度的相关性系数为 0.82。对于 $PM_{2.5}$ ，DustTrak 的逐时浓度是 TEOM 的 2.9 倍，两者浓度的相关性系数为 0.91。

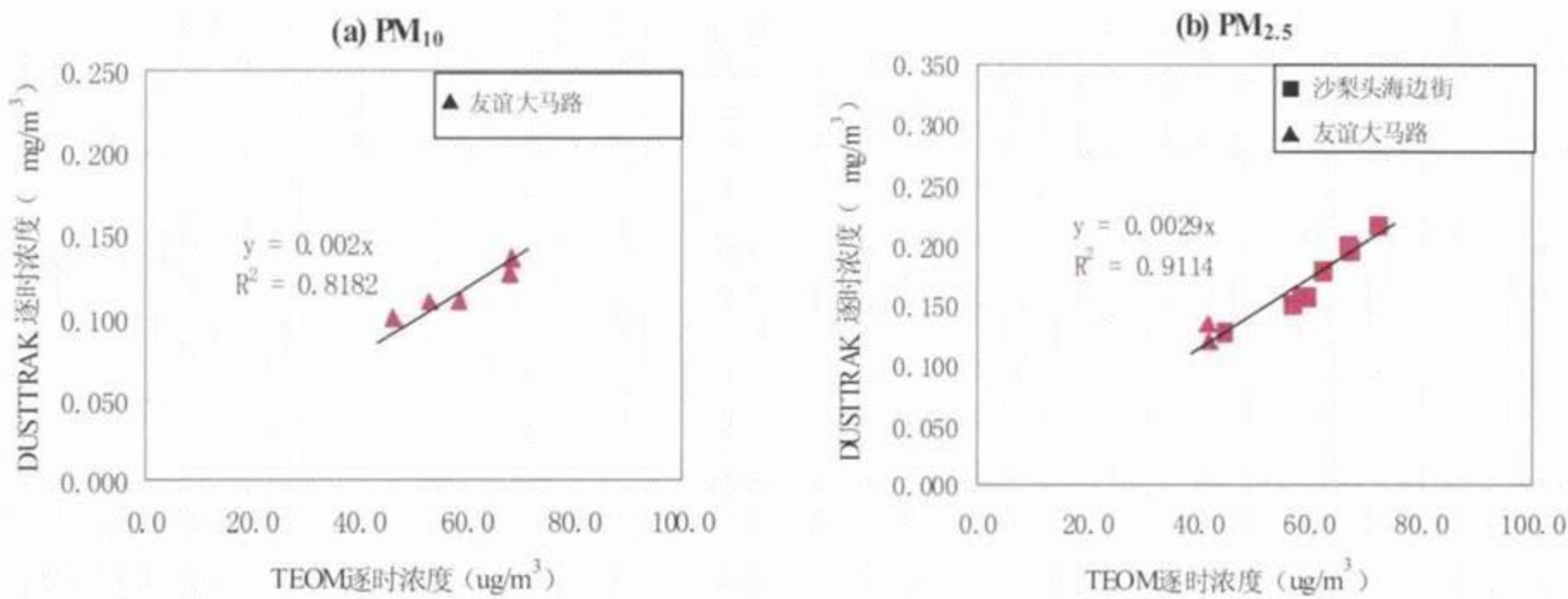


图 6 道路边 DustTrak 和 TEOM 监测仪逐时浓度的比较

由于监测期间 TEOM1400a 未配备 $1\mu m$ 的采样头，因此无法给出校正 DustTrak 仪器 PM_{10} 浓度的准确值。本研究假设 DustTrak 的 PM_{10} 浓度的校正系数与 $PM_{2.5}$ 相同，由此获得的 $PM_{2.5}$ 和 PM_{10} 在 PM_{10} 浓度中的比例见表 2。

结果表明，友谊大马路和沙梨头海边街细粒子和亚微米级粒子在 PM_{10} 中占据了很大的比例。 $PM_{2.5}$ 在 PM_{10} 中所占的份额达到了 66%-67%，而 PM_{10} 在 PM_{10} 中所占的份额也高达 51%-60%。由于澳门地处南部沿海，道路两旁裸露地面基本被植被覆盖，道路清洁，因此道路扬尘并不十分突出；此外，由于澳门街道机动车密度非常大，部分扬尘粗颗粒在车流的碾压作用下转变为较细的颗粒，这部分细颗粒与澳门道路很高的机动车尾气管细微颗粒物排放的协同作用，是导致澳门交通环境细粒子和亚微米级粒子在 PM_{10} 中比例偏高的主要原因。

表 2 澳门道路边颗粒物浓度的粒径分布

监测点	友谊大马路				沙梨头海边街			
	点 1		点 2		点 1		点 2	
粒径分布	$\frac{PM_{2.5}}{PM_{10}}$	$\frac{PM_1}{PM_{10}}$	$\frac{PM_{2.5}}{PM_{10}}$	$\frac{PM_1}{PM_{10}}$	$\frac{PM_{2.5}}{PM_{10}}$	$\frac{PM_1}{PM_{10}}$	$\frac{PM_{2.5}}{PM_{10}}$	$\frac{PM_1}{PM_{10}}$
$\overline{R_r}^a$	0.656	0.575 ^d	0.669	0.549 ^d	0.665	0.597 ^d	NA ^e	0.509 ^d
σ_r^b	0.009	0.013	0.011	0.011	0.053	0.014	NA ^e	0.049
n ^c	11	11	10	12	9	10	NA ^e	10

^a $\overline{R_r}$ = $PM_{2.5}$ 和 PM_1 在 PM_{10} 中的平均比例, 已经经过 TEOM 的校正
^b σ_r = 逐时平均浓度比值的样本方差
^c n = 逐时平均浓度比值的样本数
^d 假设 DustTrak 的 PM_1 浓度的校正系数与 $PM_{2.5}$ 相同, 为 0.0029 (见图 6)
^e NA = 由于 D6 在监测期间操作失误, 导致数据丢失

2. 道路边颗粒物的化学成分分析

2.1. 监测方法和程序

本研究选取澳门的 3 条典型道路, 进行了颗粒物 (PM_{10} 和 $PM_{2.5}$) 化学成分的采样分析, 监测地点如下: 1) 水坑尾街; 2) 友谊大马路; 3) 东望洋街。同时, 为了分析比较, 还选择了澳大附小 (城市居住区) 和大潭山 (区域背景点) 进行了 PM_{10} 和 $PM_{2.5}$ 的采样分析。

2.1.1. 监测仪器

1) TEOM

有关 TEOM 仪器的说明见 1.1.1. 的相关内容。

2) ACCU

ACCU 是美国 Rupprecht & Patashnik 公司制造的 8 通道颗粒物辅助采样系统, 与 TEOM1400a 配套使用。TEOM1400a 采样流量为 16.7L/min, 其中主流量 3.0 L/min 进入 TEOM 传感单元, 而辅流量 13.7 L/min 则通过外接管路进入 ACCU。8 通道可以设置采样时段进行自动连续采样, 但同一时间辅流量只能通过一个通道, 不能并行采集。采集颗粒物的滤膜共两种: 47mm 的 PTFE Teflon 膜 (Whatman Inc., Clifton, NJ, USA, and Gelman Inc., Ann Arbor, MI, USA) 用于元素和阴阳离子分析; 47mm 的石英膜 (Whatman Inc., Clifton, NJ, USA, and Gelman Inc., Ann Arbor, MI, USA) 用于 OC/EC 分析。

2.1.2. 监测程序

交通环境颗粒物 (PM_{10} 和 $PM_{2.5}$) 化学成分的采样地点设置在水坑尾街、友谊大马路和东望洋街。TEOM+ACCU 采样系统距离道路边约 1~2 m, 采样高度约为 3.5 m。对照点设置在澳大附小 (城市居住区) 和大潭山 (区域背景点), 采样高度约为 3.5 m。采样时间为 2001 年 5 月 8-9 日和 2001 年 7 月 18 日-8 月 13 日。对于每一张滤膜, 采样时段为 10-24 h 不等。采集完颗粒物的滤膜 0-4°C 低温冷藏。

2.1.3. 分析方法

采集完颗粒物的 Teflon 膜首先在超净室中经过精度为百万分之一的天平的称重分析, 与 TEOM 的同步监测浓度进行比较。然后将 Teflon 膜用于元素 (Ca、Si、Pb、S、Al、Fe 等 19 种元素) 和阴阳离子 (SO_4^{2-} 、 NO_3^- 、 NH_4^+ 等) 分析。元素分析采用 X 荧光法 (X-Ray Fluorescence, XRF); 阴阳离子采用离子色谱分析法 (Ion Chromatography, IC)。石英膜用于 OC/EC 分析,

分析方法采用热/光反射法 (Thermal/Optical Reflectance, TOR)。

对空白膜也分别进行上述的化学分析, 以确保在数据分析中消除空白膜的影响。

2.2. 交通环境颗粒物元素和离子成分分析

友谊大马路和东望洋街 PM_{10} 和 $PM_{2.5}$ 元素和离子成分的分析结果见图 7。主要结论如下: 1) SO_4^{2-} 、 NO_3^- 、 NH_4^+ 含量很高, 尤其是 SO_4^{2-} , 在 PM_{10} 和 $PM_{2.5}$ 中浓度都超过了 $3\mu g/m^3$ 。将 SO_4^{2-} 浓度与元素分析中的 S 含量进行比较, 发现在 PM_{10} 中, 83% 的 S 为可溶性的 SO_4^{2-} ; 而在 PM_{10} 中, 这一比例上升到了 96%。 NH_4^+ 通常被认为是中和 SO_4^{2-} 和 NO_3^- 最主要的物质, 分析结果表明, 道路边 70% 的 SO_4^{2-} 和 NO_3^- 被 NH_4^+ 中和。2) Al、Si、Ca、Fe、K、Mg 等地壳元素在 PM_{10} 中含量也比较高, 其浓度介于 $0.2-0.8\mu g/m^3$ 之间; 在 $PM_{2.5}$ 中, 这些元素的浓度略有下降, 但并不明显, 其浓度介于 $0.1-0.5\mu g/m^3$ 之间。3) 在 PM_{10} 中, Na 和 Cl 的浓度很高, 达到了 $0.8-2.1\mu g/m^3$; 而在 $PM_{2.5}$ 中, Na 和 Cl 的含量则显著下降, 浓度介于 $0.1-0.5\mu g/m^3$ 之间。Na、Cl 通常被认为是海盐的标志性元素。这是由澳门特定的地理位置造成的: 由于澳门位于南部沿海, 因此颗粒物受到了海盐成分的显著影响。4) Pb 的含量很低, 在 PM_{10} 和 $PM_{2.5}$ 中浓度仅为 $0.03\mu g/m^3$ 左右。由于自 1995 年以来, 澳门大力推行了汽油无铅化的进程, 导致大气中颗粒物的铅浓度大幅度的降低。

作为比较的澳大附小 (居住区) 和大潭山 (背景点) PM_{10} 和 $PM_{2.5}$ 元素和离子成分的分析结果见图 8。从图中可以看出, SO_4^{2-} 、 NO_3^- 、 NH_4^+ 含量依然很高, 与道路边的浓度水平相当。由于 SO_4^{2-} 、 NO_3^- 、 NH_4^+ 主要是二次反应生成的产物, 其来源很可能更多的受到较大尺度的珠三角地区的影响, 澳门当地源的影响反而是次要的, 因此, 在澳门地区的小尺度范围内, SO_4^{2-} 、 NO_3^- 、 NH_4^+ 浓度基本保持稳定。此外, 与道路边相比, 澳大附小和大潭山的 Al、Si、Ca、Fe、K、Mg 等地壳元素的浓度有所下降, 但是仍然占据一定的份额。由于澳门平均风速较大, 局部风吹尘对采样点的影响还是比较明显。

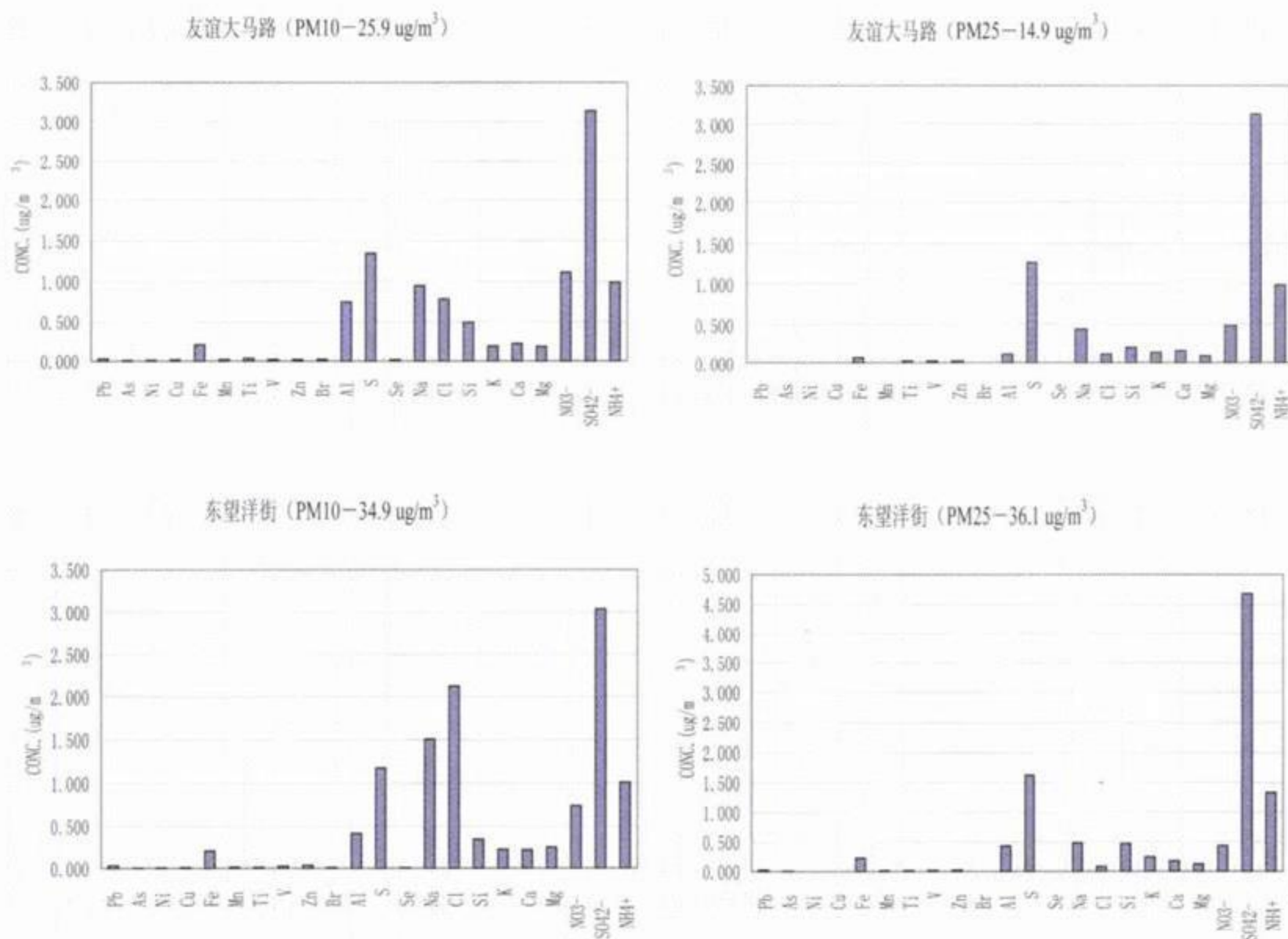


图 7 友谊大马路和东望洋街 PM_{10} 和 $PM_{2.5}$ 元素和离子成分

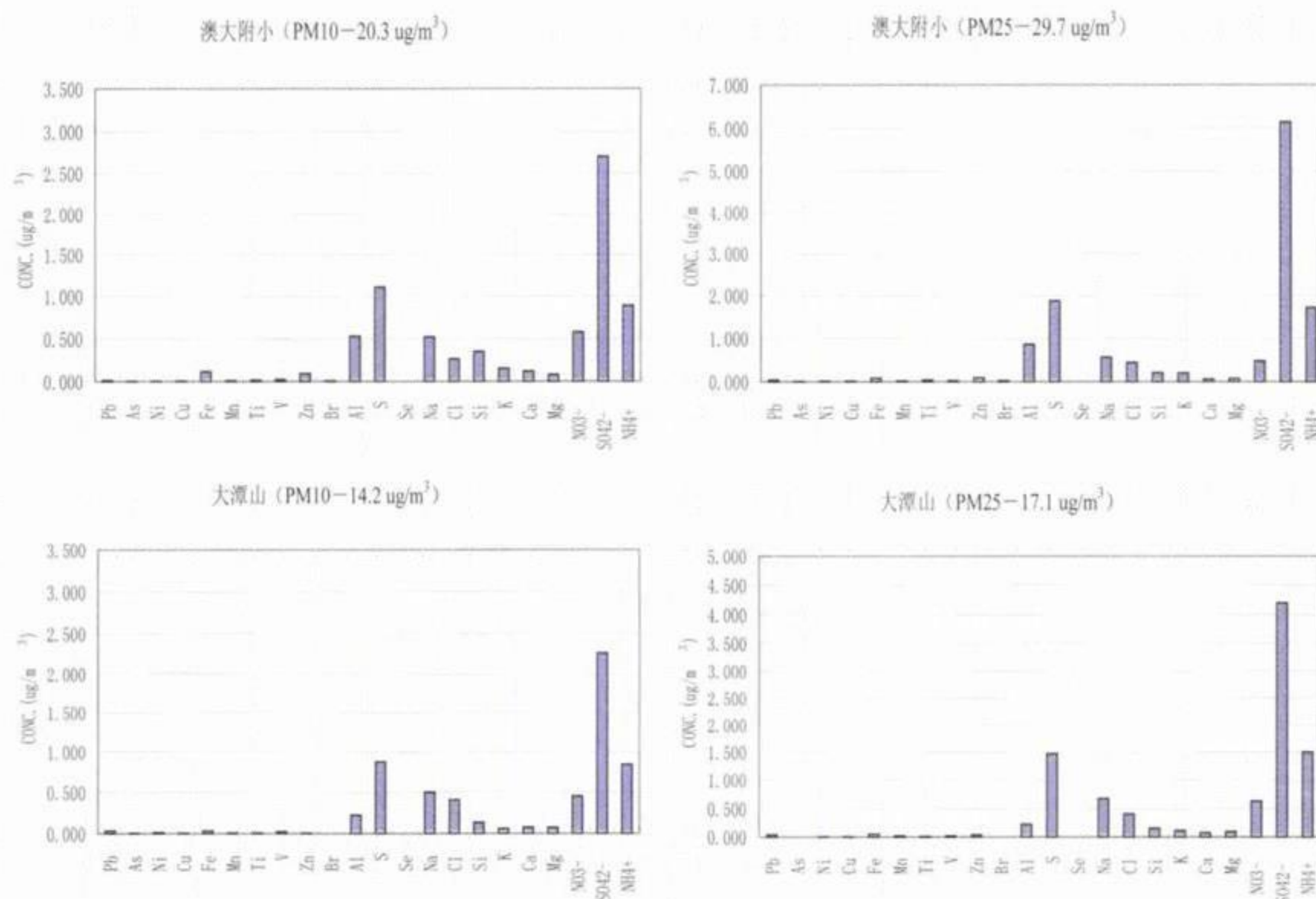


图 8 澳大附小（居住区）和大潭山（背景点）PM₁₀ 和 PM_{2.5} 元素和离子成分

2.3. 交通环境颗粒物碳成分分析

澳门道路边（友谊大马路和东望洋街）、居住区（澳大附小）和背景点（大潭山）PM_{2.5} 中的碳（OC/EC）成分分析结果见图 9。

可以看出，道路边总碳（TC）的浓度很高。友谊大马路 TC 浓度 13.0µg/m³，占采样期间 TEOM 监测浓度的 59.1%，其中 OC 和 EC 分别为 7.9µg/m³ 和 5.1µg/m³；而东望洋街 TC 浓度 15.9µg/m³，占采样期间 TEOM 监测浓度的 50.6%，其中 OC 和 EC 分别为 12.2µg/m³ 和 3.7µg/m³。

而在作为居住区和背景点的澳大附小和大潭山，TC 总浓度出现了显著的下降。澳大附小 TC 浓度 7.3µg/m³，占采样期间 TEOM 监测浓度的 34.4%，其中 OC 和 EC 分别为 6.6µg/m³ 和 0.7µg/m³；而大潭山 TC 浓度 4.0µg/m³，占采样期间 TEOM 监测浓度的 37.4%，其中 OC 和 EC 分别为 3.8µg/m³ 和 0.2µg/m³。可以看出，居住区和背景点 TC 比例的下降，更大程度上应归因于 EC 浓度的急剧减少。在道路边，OC/EC 的比值约为 3.0，到了居住区，OC/EC 比例上升到 9.4，而在作为背景点的大潭山，这一比例更高达 15.5。

EC 通常被认为是机动车，特别是柴油车排放的标志性元素^[10,11]。对采样期间典型道路的车流构成比例的统计分析，显示友谊大马路柴油车（包括出租车、公共汽车和重型柴油货车）在总车流量中的比例超过了 30%，而在东望洋街，这一比例仅为 15%。因此，友谊大马路 EC 的比例显著高于东望洋街。由于居住区受车流的影响较少，导致 EC 浓度急剧下降；而在城市背景点，EC 浓度更低，仅为 0.2µg/m³，该浓度值可以认为是 EC 的本底浓度。

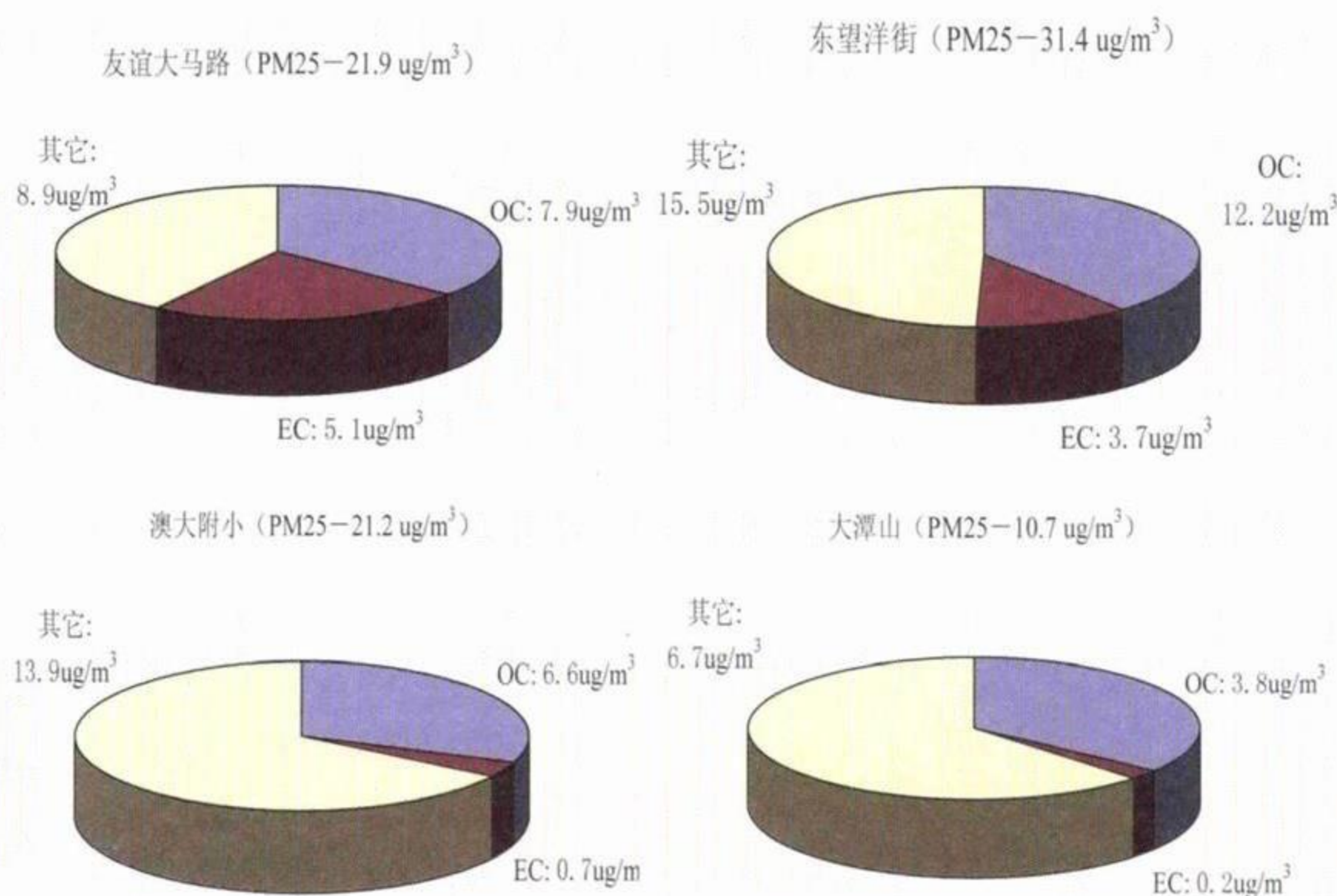


图 9 澳门道路边、居住区和背景点 PM_{2.5} 碳 (OC/EC) 成分

2.4. 交通环境颗粒物化学物质平衡分析

通过对交通环境颗粒物的化学物质平衡分析,有助于确定不同化学成分在颗粒物中的比例,并进一步定性分析各种来源贡献的大小。本研究中,颗粒物化学物质平衡分析的内容包括以下几个方面的内容:1) 有机物质 (Organic Material): 包括了数以百计的复杂的有机物,实际计算时通常在 OC 的基础上乘以某一系数。国内外多数研究认为这一系数的取值范围为 1.2-1.4 之间^[12,13],本研究取值 1.2; 2) EC; 3) 二次粒子: 以 SO_4^{2-} 、 NO_3^- 和 NH_4^+ 为代表; 5) 海盐成分: 以 Na 和 Cl 为代表; 6) 地壳物质 (Crustal): 包括了 Ca、Si、Al、Fe、Mn 等多种元素,目前多以这些元素的氧化物形式来确定地壳成分的贡献^[12-14]。本研究中,取 Ca、Si、Al、Fe、Mn、K、Mg、Ti 8 种元素各自的氧化物形式作为地壳物质; 7) 痕量组分 (Trace Species): 除地壳元素、S、海盐成分之外的其它 XRF 分析的元素总和。

道路边 PM₁₀ 和 PM_{2.5} 化学物质平衡分析见图 10。由于水坑尾街和东望洋街彼此毗邻,水平距离在 50 m 以内,可以近似认为两地颗粒物的来源相同。结果表明: 1) 有机物质是道路边颗粒物中最重要的化学成分,在 PM_{2.5} 中这一比例高达 42%,而在 PM₁₀ 中其贡献也超过了 30%。有机物的来源非常复杂,既包括了本地源的贡献,如机动车尾气管排放、道路尘、轮胎磨损/刹车板磨损、油烟等等,也包含了更大尺度区域源的重要影响,这主要是指由于光化学反应等产生的二次有机粒子。2) 这种更大尺度区域源的重要影响,从 SO_4^{2-} 、 NO_3^- 、 NH_4^+ 等二次粒子所占的比例中也得到了体现: SO_4^{2-} 、 NO_3^- 和 NH_4^+ 在 PM₁₀ 和 PM_{2.5} 中均占据了重要份额 (16-18%), 其中 SO_4^{2-} 占到了绝大部分。由于澳门本地基本没有工业排放,因此这些二次粒子主要受到了华南珠三角地区等地的影响。3) EC 主要反映了机动车尤其是柴油车排放的贡献,在 PM₁₀ 和 PM_{2.5} 中这一比例分别为 4% 和 11%。在 PM_{2.5} 中 EC 比例显著升高,这主要是因为柴油车排放的 EC 基本由细粒子组成。4) 地壳元素在 PM₁₀ 和 PM_{2.5} 中所占的份额分别为 10% 和 8%,这表明道路扬尘对颗粒物有重要的影响。与 PM₁₀ 相比,地壳元素在 PM_{2.5} 中的比例仅略微的下降。在澳门,道路尘中细粒子很可能占据了很大的份额,粗粒子比例并不高,这与道路边粒径分布的监测结果是相吻合的。5) 澳门由于受到了海洋的强烈影响,Na 和 Cl

的比例比较显著。从图中可以发现，在 PM_{10} 中这一比例显著高于 $PM_{2.5}$ ，这表明澳门区域海盐粒子中粗粒子比重比较大。

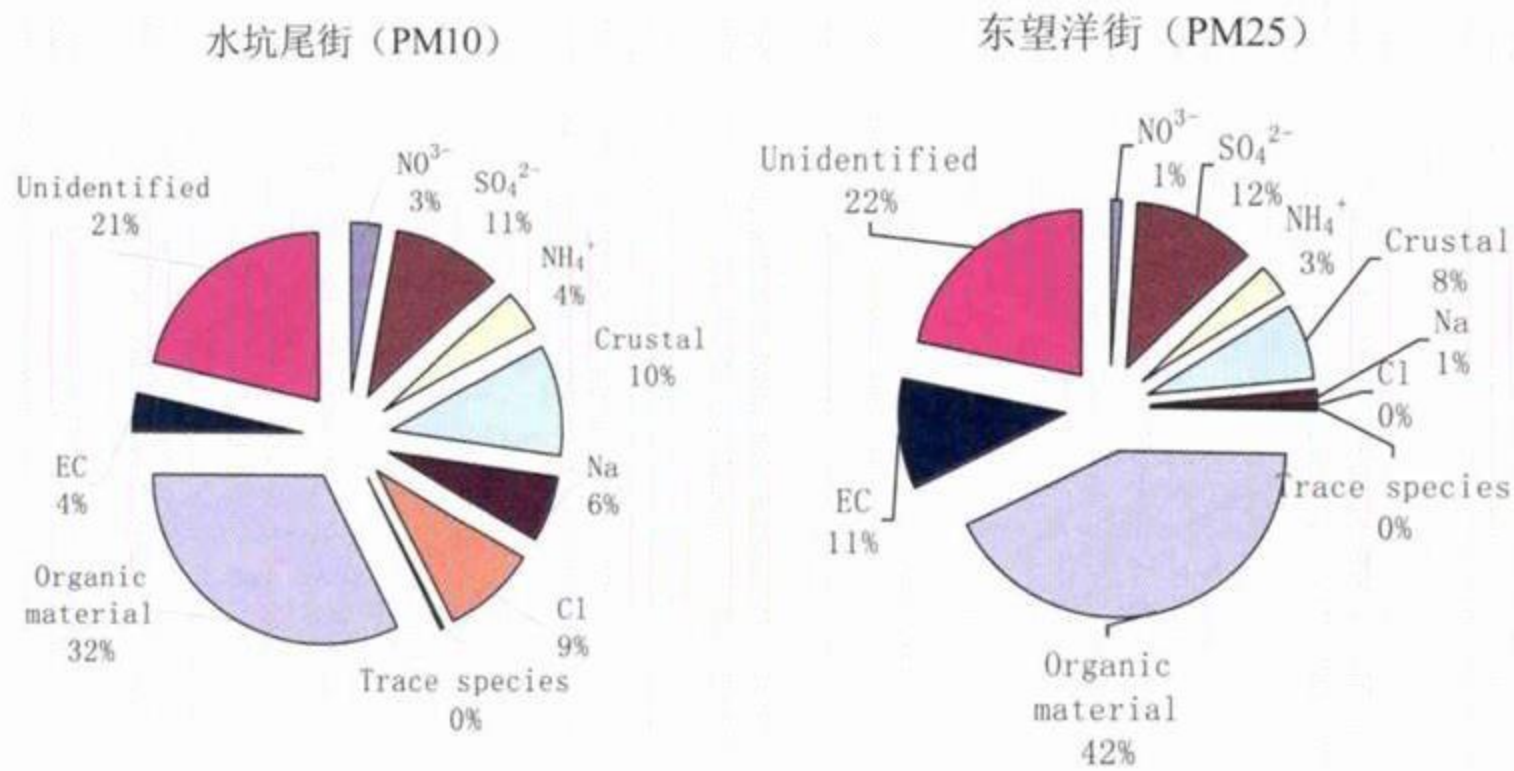


图 10 澳门道路边 PM_{10} 和 $PM_{2.5}$ 化学物质平衡分析

澳门居住区和背景点的 $PM_{2.5}$ 化学物质平衡分析见图 11。与道路边的相比，其化学物质的比例有所不同：1) 有机物质依然是最重要的化学物质，在居住区和背景点的比例依然高达 33% 和 38%，虽然该比例与道路边的相比略有下降。由于居住区和背景点受道路源（机动车排放等）的影响要小得多，因此这部分有机物质应主要来自于更大尺度的区域源，例如由于光化学反应等产生的二次有机粒子。2) 与道路边相比，EC 比例大幅度下降，这是由于机动车排放对居住区和背景点的影响急剧减少造成的。3) SO_4^{2-} 、 NO_3^- 、 NH_4^+ 等二次粒子所占的比例显著上升，在居住区和背景点的比例分别达到了 24% 和 34%。这也主要归因于居住区和背景点受道路源（机动车排放等）等本地源的影响逐渐减小，从而导致更大尺度区域背景源的影响比例显著上升。4) 澳大附小和大潭山的地壳元素的比例仍然占据一定的份额（10% 左右）。这表明局部风吹尘对采样点的影响还是比较明显。

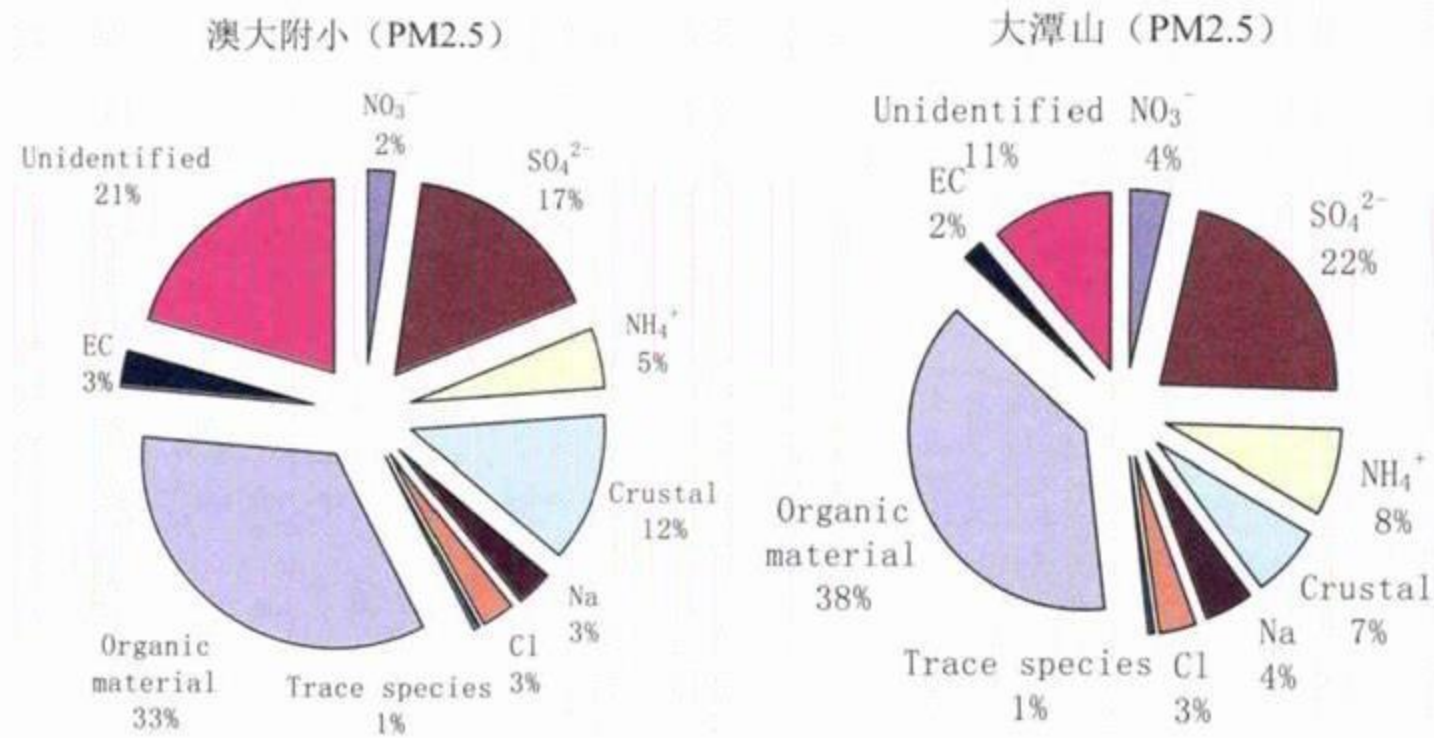


图 11 澳门居住区和背景点 $PM_{2.5}$ 化学物质平衡分析

3. 结论

通过现场监测和采样，对澳门交通环境颗粒物基本的物理和化学特征进行了分析。获得了道路边颗粒物浓度的空间分布、粒径分布和主要的化学成分谱。

采用 DustTrak 和 TEOM1400a 颗粒物监测仪对道路边颗粒物浓度的垂直分布、水平分布和

粒径分布进行了监测。结果表明: 1) PM_{10} 、 $PM_{2.5}$ 和 PM_1 的浓度均表现出了明显的随高度增加而递减的规律, 在 79m 处, PM_{10} 、 $PM_{2.5}$ 和 PM_1 的浓度分别衰减为地面最大浓度的 60%、62% 和 80%; 2) 道路边颗粒物水平方向上的衰减趋势并不明显, 在距道路边 0-228m 范围内, PM_{10} 、 $PM_{2.5}$ 和 PM_1 的最大浓度衰减幅度仅分别为 7%、9% 和 10%; 3) 道路边 $PM_{2.5}$ 和 PM_1 分别贡献了 PM_{10} 总浓度的 66-67% 和 51-60%, 表明澳门道路边细粒子和亚微米级粒子占据了 PM_{10} 的大部分份额。

采用 TEOM1400a 和 ACCU 颗粒物系统对澳门道路边 PM_{10} 和 $PM_{2.5}$ 进行了采样, 并进行了化学成分分析。结果表明: 1) 有机物质是道路边颗粒物中最重要的成分, 无论是在 PM_{10} 还是 $PM_{2.5}$ 中, 其比例都超过 30%; 2) SO_4^{2-} 等二次粒子、EC、地壳元素和海盐成分也是颗粒物的重要组成部分, 分别占 5-20% 不等的份额; 3) 在城市居住区和背景点 EC 浓度很低, 但在道路边, 特别是柴油车流量较大的交通环境, EC 比例很高, 这表明 EC 是反映机动车尤其是柴油车排放的标志性元素; 4) 初步分析结果显示道路边的 $PM_{10}/PM_{2.5}$ 来源比较复杂, 机动车排放、道路尘等可能是重要的污染源, 此外二次例子和背景浓度等较大尺度的区域性外来源也是很重要的影响因素。

4. 致谢

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Physical and chemical characteristics of airborne particulate matter near major roads in Macao, China

Hao Jiming, Wu Ye, Fu Lixin, Hu Jingnan, Wang Litao

(Department of Environmental Science and Engineering, Tsinghua University, Beijing 100084)

Wang Zhishi, Tang U Wa

(Faculty of Science and Technology, University of Macao, Macao)

Abstract: A monitoring study on physical and chemical characteristics of airborne particulate matter was conducted near major roads in Macao. Vertical profiles, horizontal profiles and size distribution of airborne particulate matter were measured using DustTrak and TEOM monitors. The essential results show as follows: 1) A significant decrease in the concentrations of particulate matter, as the height above the ground increases from 2m to 79m, was found. At the height of 79m, the concentrations of PM_{10} , $PM_{2.5}$ and PM_1 , decrease to about 60%, 62% and 80% of the maximum occurring at 2m above the ground, respectively. 2) The horizontal profiles near another major road reveal there is no significant trend of decrease in concentrations of particulate matter as the distance from the road increases. Over the total measured distance (0-228m), the maximum decreases of PM_{10} , $PM_{2.5}$ and PM_1 are only 7%, 9% and 10%, of the maximum occurring at 2m from the road, respectively. 3) The daytime averaged $PM_{2.5}$ and PM_1 contribute 66%-67% and 51%-60%, respectively, of the total PM_{10} mass after the particle readings by DustTrak were recalibrated by TEOM. It shows that fine particles and submicrometer particles contribute major part of PM_{10} at the roadside in Macao. PM_{10} and $PM_{2.5}$ were sampled using TEOM and ACCU system, and then the chemical composition was analyzed. The essential results show as follows: 1) Organic material is the most abundant species, constituting more than 30% of the total PM_{10} and $PM_{2.5}$ at all sites. 2) The secondary species including SO_4^{2-} , NO_3^- and NH_4^+ , elemental carbon, crustal species, and sea salt (NaCl) are also important components, accounting for 5-20%, respectively, of the PM_{10} and $PM_{2.5}$ mass. 3) Elemental carbon near major roads, especially the road having more diesel vehicles on it, is much higher than those sites in resident and background areas. It suggested that EC could be considered as the mark species for vehicles, especially the diesel vehicles. 4) The sources for roadside PM_{10} and $PM_{2.5}$ are complicated, which most likely include vehicle tailpipe exhausts, paved road dust, residential cooking, and other regional-scale sources.

Keywords: Particulate matter; Vertical profiles; Horizontal profiles; Particle size distribution; Chemical composition; Traffic

澳门机动车尾气排放特征及污染影响研究

郝吉明 胡京南 吴烨 傅立新

(清华大学环境科学与工程系)

王志石 邓宇华

(澳门大学科技学院)

摘要: 机动车尾气排放是澳门地区主要的大气污染源,对当地的空气质量产生重要影响,因而有必要对澳门机动车的尾气排放特征及其影响开展研究。本文中的双怠速排放检测是在2000-2001年间分两次于澳门车检所内进行,其中汽油轿车和轻型客车样本共37辆,摩托车样本为20辆。同时,利用AVL五气分析仪,还对七辆汽油轿车样本进行了实际运行工况下的尾气排放随车监测。结果表明,对于汽油轿车,无论是双怠速还是实际运行工况,化油器车(主要是1995年以前注册的旧车)在CO、HC和NO_x排放上都明显高于安装电喷加三元催化设备的新车。怠速时两者CO排放的体积浓度平均值分别为2.73%和0.01%,HC分别为360ppm和54ppm(按正己烷当量);实际运行工况下化油器车NO_x排放的体积浓度均值为943ppm,而后者则为140ppm。由于油温较低,催化设备在实际运行循环的启动阶段容易出现失效。二冲程和四冲程摩托车的HC排放水平存在很大差别,前者怠速状态下HC排放的体积浓度平均值为5,957ppm(按正己烷当量),远大于四冲程摩托车的238ppm。此外,在分析澳门城区的机动车排放污染影响时,通过比较三个自动监测站点的数据,发现交通干道边的NO_x浓度比居民区和背景点高一倍以上,PM₁₀浓度也偏高。对该道路的车流量和路边的污染物浓度监测数据进行时变化相关性分析,结果表明车流量和PM₁₀、NO₂、NO和CO浓度之间都呈现良好的相关性,线性回归得到的R²值在0.6-0.9之间。

关键词: 尾气排放检测 双怠速 实际运行工况 相关性

1. 引言

位于珠江三角洲西南部的澳门,面积仅23.8平方公里,人口为43.4万,人口密度近20,000人/平方公里,且90%以上的人口都居住在面积为7.8平方公里的澳门半岛。90年代以来,澳门的机动车保有量以年均9.8%的比率快速增长,截至1999年底,已达到11.3万辆,是世界上机动车密度最高的地区之一^[1]。机动车排放是澳门最主要的大气污染源,其1999年排放的CO、HC、NO_x、PM₁₀和PM_{2.5}分别为14251.2、2195.3、2408.2、140.0和127.5吨^[2]。根据何东全等人的研究,澳门地区主要的交通干道周围有出现机动车排放严重污染的可能性^[3]。

鉴于对当地空气质量产生的重要影响,有必要对澳门机动车的尾气排放特征及其影响开展研究。本研究通过对37辆汽油轿车和轻型客车样本的双怠速排放测试和20辆摩托车样本的怠速排放测试,以及七辆汽油轿车样本在实际运行工况下的尾气排放随车监测,获得了基于不同车龄、控制技术和不同运行工况下机动车污染物的排放规律。此外,还比较了三个空气质量自动监测站的数据,并对交通干道的车流量和道路边的污染物浓度进行了时变化相关性分析。

2. 实验方法

2.1. 双怠速排放测试

研究采用 AVL 五气分析仪对澳门车辆检测场的 37 辆汽油轿车和轻型客车样本进行了双怠速污染物排放检测，同时对 20 辆摩托车样本进行了怠速污染物排放检测，受检车的状况见表 1，代表了不同车龄和类别的车辆。怠速排放检测的车辆发动机转速为 800rpm 左右（摩托车发动机的测试转速为 1000rpm 左右），高怠速排放检测的发动机转速为 2000rpm 左右。检测的排放污染物包括 CO、HC 和 NO_x 三种。同时还检测了与燃烧状况相关的参数，如 CO₂、O₂ 浓度和过量空气系数 λ。

表 1 受检汽油车的车辆状况

汽油轿车和轻型客车的双怠速测试					
车辆编号	1-19	20	21-30	31-35	36-37
车龄（年）	>6	3	0	10-13	4-5
车型类别	轿车	轿车	轿车	轻型客车	轻型客车
摩托车的怠速测试					
车辆编号	1-8	9	10	11-16	17-20
车龄（年）	8-10	3	0	3-6	0
车型类别	二冲程	二冲程	二冲程	四冲程	四冲程

注：车龄为0表示正在注册的新车

2.2. 实际运行工况下的排放测试

研究采用 AVL 五气分析仪对七辆汽油轿车样本进行了实际运行工况下尾气排放的随车监测，测试的污染物包括 CO、HC 和 NO_x 三种。测试的行驶路线为：从位于青洲坊北的澳门车辆检测场，经关闸，沿马场北大马路而下，在行驶至约 2 公里处的环岛（东北大马路和友谊桥大马路交界处）折返，顺原路开回车辆检测场，路线总长约 4km。车辆在道路上的行驶状况为：下坡→转弯（180 度）→上坡→下坡→平路（车速基本稳定在 50km/h）→环岛转弯（180 度）→平路（车速基本稳定在 30km/h）→上坡，车速的变化范围在 0-60km/h。每辆车进行两次平行实验，七辆车的测试循环（包括各路段的行驶速度）基本保持一致。七辆轿车的状况见表 2，代表了不同控制水平的车辆。其中车辆 A、B 和 C 的测试在 2000 年 11 月进行，每一个循环人工记录约 20 个时刻的数据；车辆 D、E、F 和 G 的测试在 2001 年 5 月进行，测试循环中每隔一秒钟自动记录一次数据。

表 2 实际运行工况测试车辆的状况

车辆编号	车龄（年）	行驶里程（km）	控制技术水平
A	1.5	5331	采用电子喷射技术和三元催化转化器
B	12	103235	化油器车，未配催化转化器
C	4	44688	采用电子喷射技术和三元催化转化器
D	4.5	50270	采用电子喷射技术和三元催化转化器
E	10	50359	化油器车，未配催化转化器
F	1.5	16814	采用电子喷射技术和三元催化转化器
G	6	40963	采用电子喷射技术和三元催化转化器

3. 实验结果和讨论

3.1. 双怠速排放测试

3.1.1. 汽油轿车和轻型客车的双怠速排放测试

图 1 和图 2 给出了不同汽油轿车和轻型客车在双怠速排放测试中得到的 CO 和 HC 浓度。测试结果表明，不同车辆的污染物排放浓度差别较大，怠速测试下 CO 和 HC 体积浓度的变化

范围分别是 0-8.49% 和 4-1356ppm（按正己烷当量），高怠速测试下则分别是 0-5.13% 和 3-2550ppm（按正己烷当量）。由于澳门自 1995 年 1 月 1 日起，要求新汽油车安装催化转化系统，其排放控制水平有了明显的提高（相当于达到了 EURO1 标准），因此在评估上述受检车辆的排放状况时，需要采用不同的排放标准。

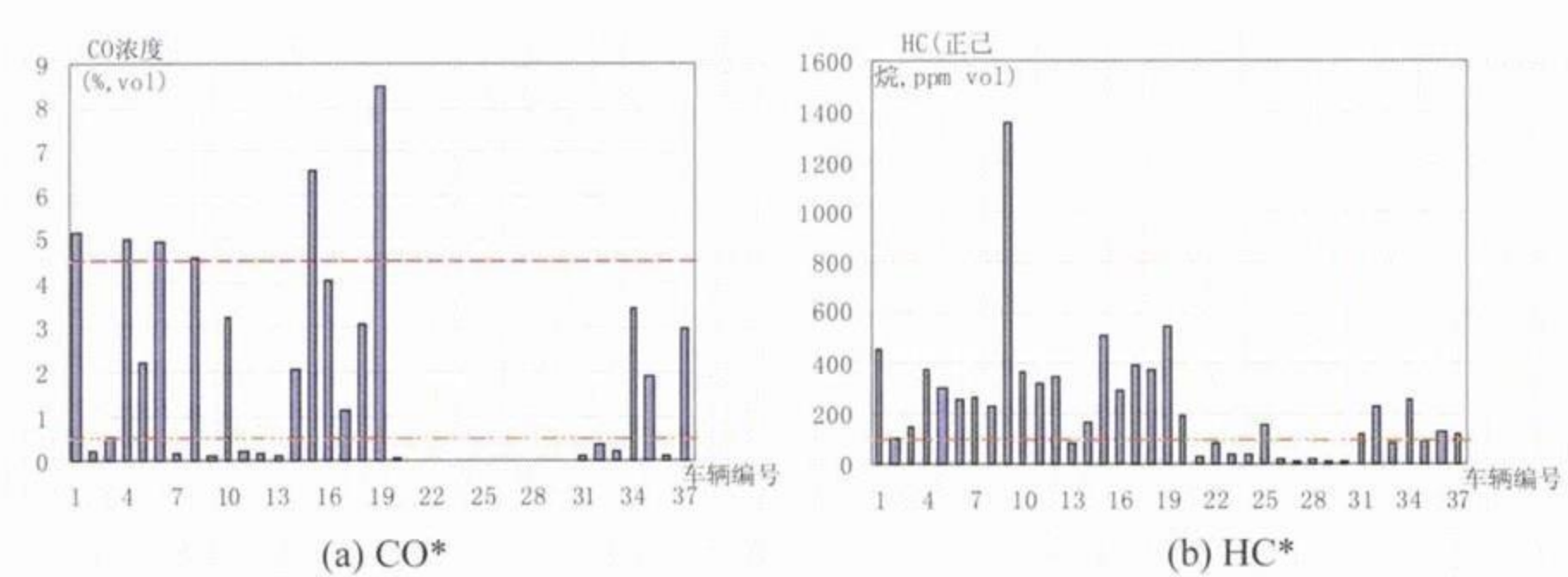


图 1 不同汽油轿车和轻型客车怠速测试的污染物排放浓度

*：CO 和 HC 排放浓度图中上下两条虚线分别代表不同的怠速参考标准，上虚线是北京市对化油器车规定的在用车怠速污染物排放标准，CO 为 4.5%，HC 为 900ppm；下虚线则是北京市对电喷车采用的怠速污染物排放标准，CO 为 0.5%，HC 为 100ppm^[4]。

本研究以北京市针对化油器车和电喷车实行的不同双怠速排放标准（如图 1 和图 2 中虚线所示）为参考，对受检车辆的排放水平进行了评估：

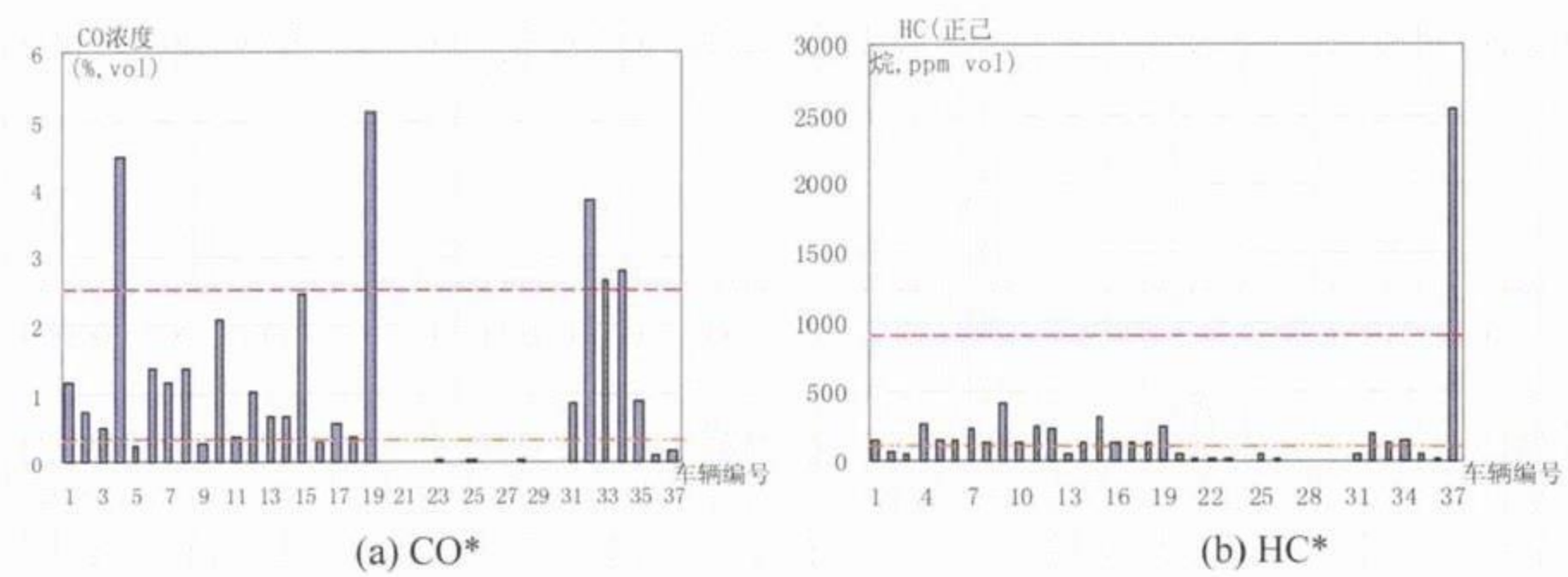


图 2 不同汽油轿车和轻型客车高怠速测试的污染物排放浓度

*：CO 和 HC 排放浓度图中上下两条虚线分别代表不同的高怠速参考标准，上虚线是北京市对化油器车规定的在用车高怠速污染物排放标准，CO 为 2.5%，HC 为 900ppm；下虚线则是北京市对电喷车采用的高怠速污染物排放标准，CO 为 0.3%，HC 为 100ppm^[4]。

- 1) 编号为 1-19 的汽油轿车（1995 年以前注册的旧车，均为化油器车），怠速和高怠速测试时 CO 排放的体积浓度平均值分别为 2.73%和 1.31%，HC 分别为 360ppm 和 178ppm（按正己烷当量）。而编号为 20-30 的汽油轿车（1995 年以后注册的在用车和新车，均为电喷车），怠速和高怠速测试时 CO 排放的体积浓度平均值均为 0.01%，HC 则分别为 54ppm 和 15ppm。虽然双怠速测试时 NOx 的排放浓度较低，但也呈现相同的趋势。说明采用了电子喷射技术和催化转化系统的车辆，排放控制水平较高，尽管在双怠速条件下催化剂性能由于油温较低等原因尚未处于较好的状态，其污染物排放浓度还是远低于化油器车。
- 2) 编号为 1-19 的汽油轿车参考北京市的化油器车双怠速排放标准。结果表明，怠速排放

- 测试中，有六辆车的排放浓度超 CO 的标准（六辆车的排放浓度范围为 4.55-8.49%），超标比例是 32%；有一辆车的排放浓度超 HC 的标准（为 1356ppm），超标比例是 5%。高怠速排放测试中，有两辆车的排放浓度超 CO 的标准（两辆车的 CO 排放浓度范围为 4.48-5.13%），超标比例 11%；HC 排放状况较好，无超标情况。
- 3) 编号为 20-30 的汽油轿车参考北京市的电喷车双怠速排放标准。结果表明，除了怠速排放测试中，有两辆车的排放浓度超 HC 的标准（两辆车排放浓度范围为 156-192ppm），超标比例是 18%；怠速排放测试中的 CO 排放浓度、高怠速排放测试中的 CO 和 HC 排放浓度均低于相应的标准。
- 4) 编号为 31-37 的轻型客车（均为化油器车），参考北京市的化油器车双怠速排放标准。结果表明，怠速排放测试中，七辆车的 CO 和 HC 排放浓度均低于相应限值，其浓度平均值分别为 1.31%和 147ppm；高怠速排放测试中，三辆车的排放浓度超 CO 的标准（三辆车的排放浓度范围为 2.66-3.83%），超标比例是 43%；有一辆车的排放浓度超 HC 的标准（为 2550ppm），超标比例是 14%。

3.1.2. 摩托车的怠速排放测试

表 3 中给出了对 20 辆摩托车排放水平进行怠速测试的数据统计结果。

表 3 摩托车怠速测试结果

编号	污染物排放浓度的变化范围			平均值		
	CO(%vol)	HC(ppmv)	NOx(ppmv)	CO(%)	HC(ppmv)	NOx(ppmv)
1-8	1.34-3.76	1955-12317	30-407	2.47	5862	124
9	4.19	8119	153	4.19	8119	153
10	2.11	4560	N/A	2.11	4560	N/A
11-16	0.08-5.28	68-463	12-63	2.37	268	39
17-20	0.10-5.77	23-398	19-76	2.47	195	46

测试结果表明：

- 1) 二冲程和四冲程摩托车的 HC 排放水平存在很大差别，前者的 HC 排放远高于四冲程摩托车。二冲程摩托车样本排放的 HC 体积浓度平均值为 5957ppm（按正己烷当量），而四冲程摩托车则为 238ppm，两者相差 25 倍。这与台湾成功大学进行的摩托车台架测试得到的结论类似^[5]。二冲程和四冲程摩托车的 CO 排放水平比较接近，但后者的 CO 排放个体差异大于二冲程摩托车，其最小值和最大值相差 50 倍以上，而二冲程摩托车仅相差 3 倍左右。二冲程摩托车的 NOx 排放稍高于四冲程摩托车。因此，采取淘汰二冲程摩托车，转向使用四冲程摩托车的措施，将能很有效地降低摩托车的总 HC 排放。
- 2) 以北京市加严的摩托车怠速排放标准为参考（适用于北京地区 2001 年以后的新注册摩托车，二冲程和四冲程摩托车的 HC 排放浓度限值分别为 3000ppm 和 300ppm）^[6]，编号为 10 的二冲程新摩托车 HC 排放超过标准限值（为 4560ppm），编号为 17-20 的四冲程新摩托车中有两辆 HC 排放超标（两辆车的排放浓度范围为 325-398ppm），超标比例是 50%。而以北京市原有的摩托车怠速排放标准为参考（适用于北京地区 2001 年以前生产的在用摩托车，二冲程和四冲程摩托车的 HC 排放浓度限值分别为 8000 ppm 和 2200ppm）^[6]，编号为 1-9 的二冲程在用摩托车中有四辆 HC 排放超标（四辆车的排放浓度范围为 8119-12317ppm），超标比例是 44%，编号为 11-16 的四冲程在用摩托车的 HC 排放则全部低于限值。
- 3) 以北京市加严的摩托车怠速排放标准为参考（CO 排放浓度限值为 1.5%）^[6]，编号为 10 的摩托车 CO 排放超标（为 2.11%），编号为 17-20 的摩托车中有两辆 CO 排放超标（两辆车的排放浓度范围为 3.89-5.77%），超标比例 50%。而以北京市原有的摩托车怠速排放标准为参考（CO 排放浓度限值为 4.5%）^[6]，编号为 1-9 的摩托车 CO 排放浓度全部低于限值，编号为 11-16 的摩托车中有一辆 CO 排放超标（为 5.28%），超标比例 17%。
- 4) 从三种污染物排放浓度的检测结果看，无论二冲程还是四冲程，新注册摩托车和在用

摩托车差别不大，有些新车的排放甚至高于多年车龄的在用车。这和成功大学的摩托车台架测试得出的结论相差很大^[5]。

3.2. 实际运行工况下的排放测试

七辆车在实际运行工况下污染物排放浓度的变化范围和两次平行实验的均值比较分别见表 4 和表 5。

表 4 测试汽油车实际行驶工况下污染物排放浓度的变化范围

车辆编号	CO (%vol)	HC (ppmv)	NOx (ppmv)
A	0-3.67	10-281	2-292
B	0.38-3.87	123-345	541-3308
C	0-0.29	2-55	0-579
D	0-2.3	4-490	0-1131
E	0.36-3.46	87-1482	129-2127
F	0-1.43	1-82	0-2007
G	0.01-7.97	10-313	6-2702

表 5 测试汽油车实际行驶工况下污染物排放浓度的平均值比较

车辆编号	测试一			测试二		
	CO (%vol)	HC (ppmv)	NOx (ppmv)	CO (%vol)	HC (ppmv)	NOx (ppmv)
A	0.51	41	37	0.24	46	70
B	1.93	210	2145	1.72	184	2098
C	0.04	20	83	0.06	20	86
D	0.10	45	44	0.03	20	66
E	1.24	241	843	1.46	300	804
F	0.06	12	91	0.05	7	121
G	0.51	56	356	N/A	N/A	N/A

注：车辆 E 和 F 的第二次试验中有部分数据未能获得，车辆 G 只进行了一次试验。

从表中可以看出：

- 1) 排放控制水平不同的车辆在实际运行工况下污染物的排放浓度差别很大。采用电喷技术和三元催化转化系统的汽油轿车（车辆 A、C、D、F 和 G）和无催化转化系统的化油器车（车辆 B 和 E）相比，前者的 CO、HC 和 NOx 排放都要低很多。电喷车 NOx 排放的体积浓度平均值为 140ppm，而化油器车则为 943ppm，约是前者的 7 倍。这说明催化转化系统在实际运行工况下发挥了很好的效用。
- 2) 比较不同车龄和行驶里程的电喷车，发现车辆 C 和 D 虽然车龄为 4 年和 4.5 年，行驶里程约 4.5 万和 5 万公里，但它们的污染排放水平和车龄均为 1.5 年，行驶里程低于 1.7 万公里的车辆 A 和 F 基本相同。这表明车辆 C 和 D 的催化转化系统在行驶 4.5 万公里之后，仍保持良好稳定的催化性能，排放劣化趋势得到很好控制，劣化水平不明显。车辆 G 的车龄为 6 年，行驶里程约 4.1 万公里，其污染物排放，特别是 NOx 的排放水平较前四辆电喷车偏高，表明其排放已开始出现劣化趋势。

图 3 和图 4 中给出的污染物排放浓度变化是两个较典型的实际运行循环样本，其横坐标为时间（单位：秒）。从图上可以看出，车辆 D 除启动阶段外，整个运行过程中三种污染物的排放都很低；而车辆 E 则自始至终保持较高的排放水平。车辆 D 的污染物排放浓度在启动阶段出现很高的峰值，反映了此时油温较低，催化转化设备发挥的效用很小。在其它的实际运行循环中，也监测到同样的情况。采取措施避免启动阶段时催化转化设备失效的发生，将有助于降低机动车的总体排放水平。

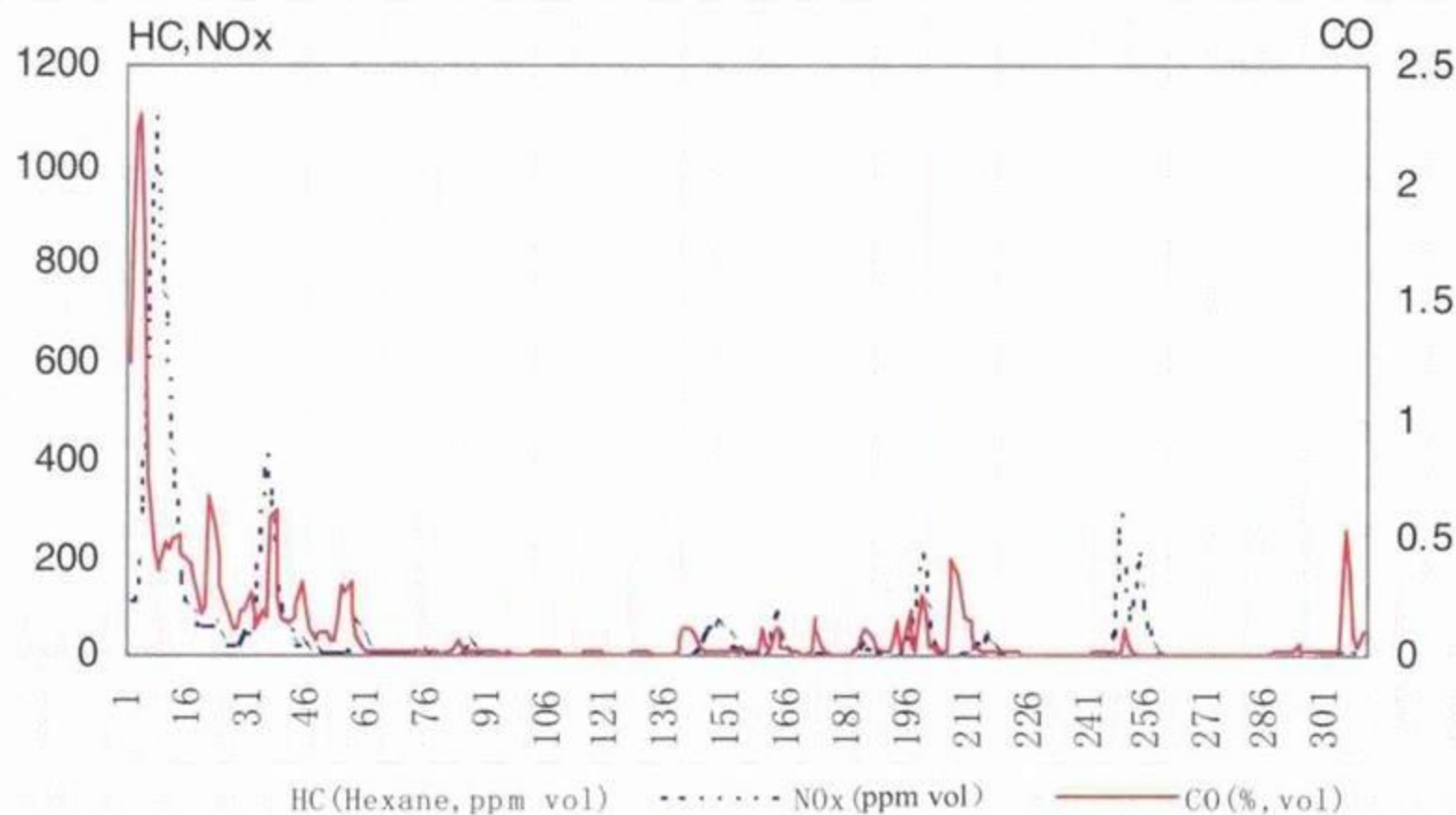


图3 车辆D在实际运行工况下污染物排放的随车监测结果

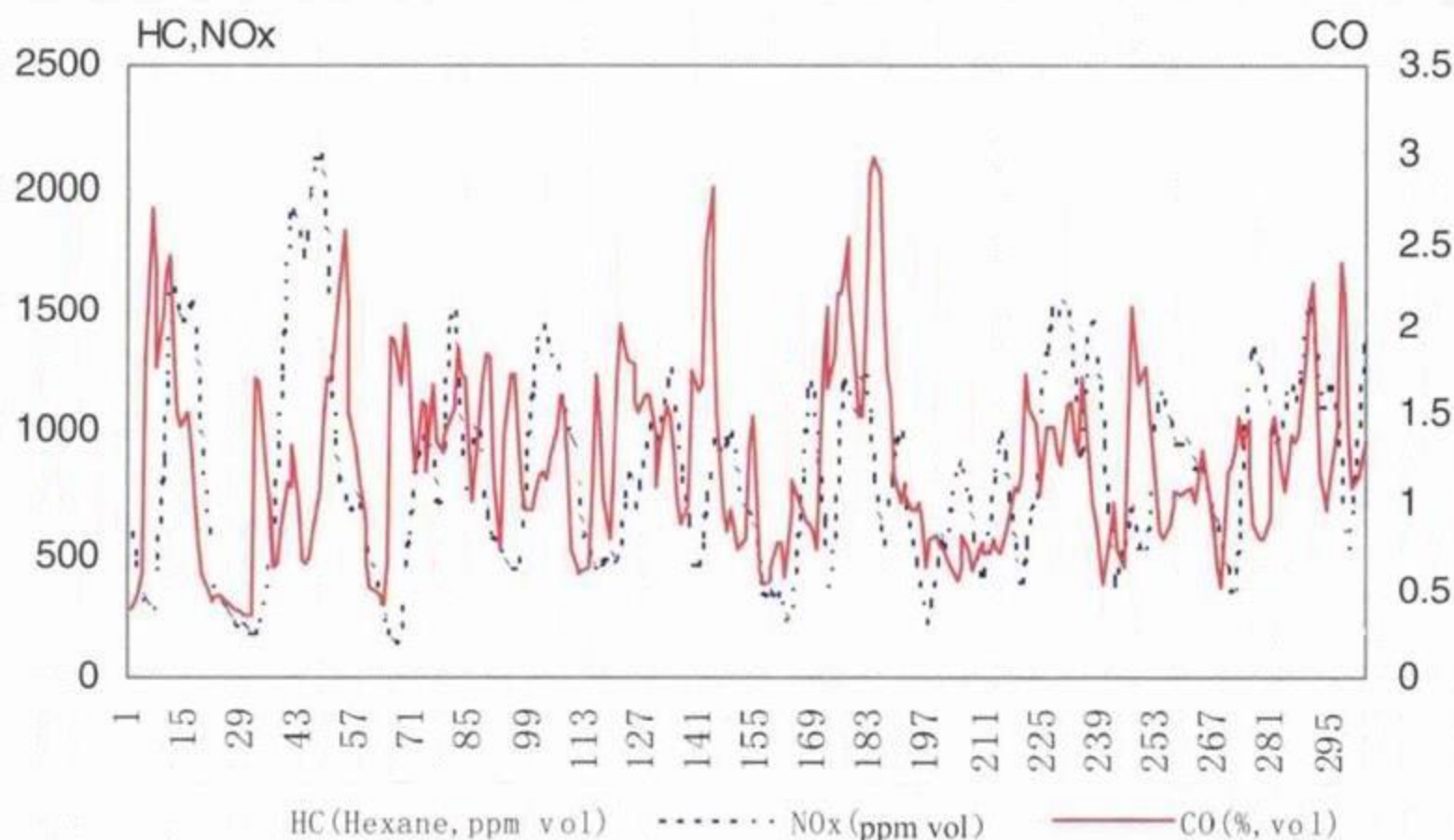


图4 车辆E在实际运行工况下污染物排放的随车监测结果

需要指出的是，上面的13次实际运行循环中，共有三个驾驶员参与了工作。有研究表明，不同的驾驶方式（如加速的强度和持续时间）对机动车的CO和NOx排放水平有着重要的影响^[7]。但本研究在对监测结果进行比较时，未考虑因驾驶员不同而可能造成的排放水平差异。此外，从图上还能观察到，CO和NOx的排放浓度峰值之间似乎存在一定的关联。两者很少同时发生，一般有一个时间延迟，但每个CO浓度峰值的附近，几乎都有一个NOx浓度峰值。分析其原因，可能是实际运行工况下行驶速度不稳定，加减速交替进行导致间隔出现CO和NOx的浓度峰值。

4. 机动车排放污染影响研究

4.1. 空气质量监测数据分析

根据从澳门地球物理暨气象局的三个自动监测站获得的数据，对从 2000 年 10 月 1 日—2001 年 9 月 31 日澳门地区不同地点的空气质量进行了比较和分析，其中包括典型道路边监测点（水坑尾），背景点（大潭山）和一般居住区监测点（化验所）。数据统计的结果见图 5，其中 NO_x 和 PM_{10} 的浓度是这一年内小时浓度的平均值。

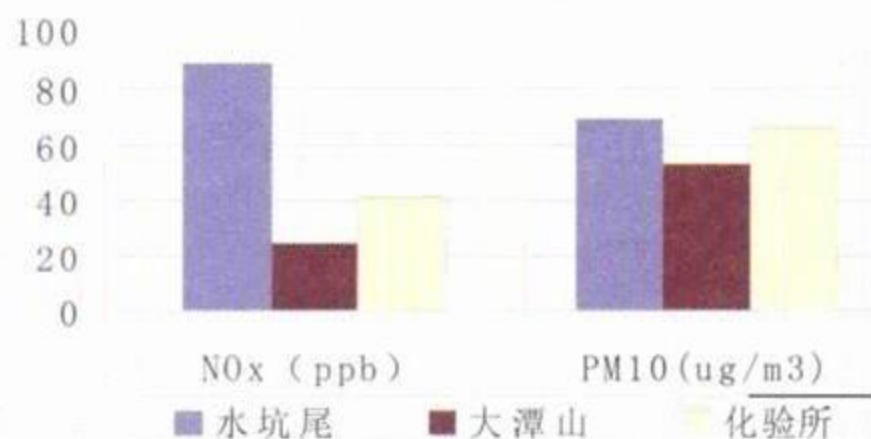


图 5 澳门地区不同地点空气质量监测数据

从图上可以看到，水坑尾站的 NO_x 监测浓度比其它两个监测站的数据高一倍以上，说明机动车排放对环境空气质量有着显著的影响。 PM_{10} 的监测浓度也偏高，但不如 NO_x 的差距明显，一方面因为当地环境中 PM_{10} 的背景浓度较高，另一方面由于 PM_{10} 的人为源很多，如油烟和工业粉尘等，机动车造成的排放只是其中的一部分。

4.2. 交通干道车流量和大气污染物浓度的相关性分析

根据位于水坑尾街的路边自动监测站数据，找出从 1999 年 8 月到 2001 年 10 月间，该监测点处于街道下风向位置的一共 36 天进行统计，得到几种污染物时均值与车流量时变化的相关性分析，结果如图 6 所示，其中左纵坐标为污染物时均浓度，右纵坐标为小时车流量。

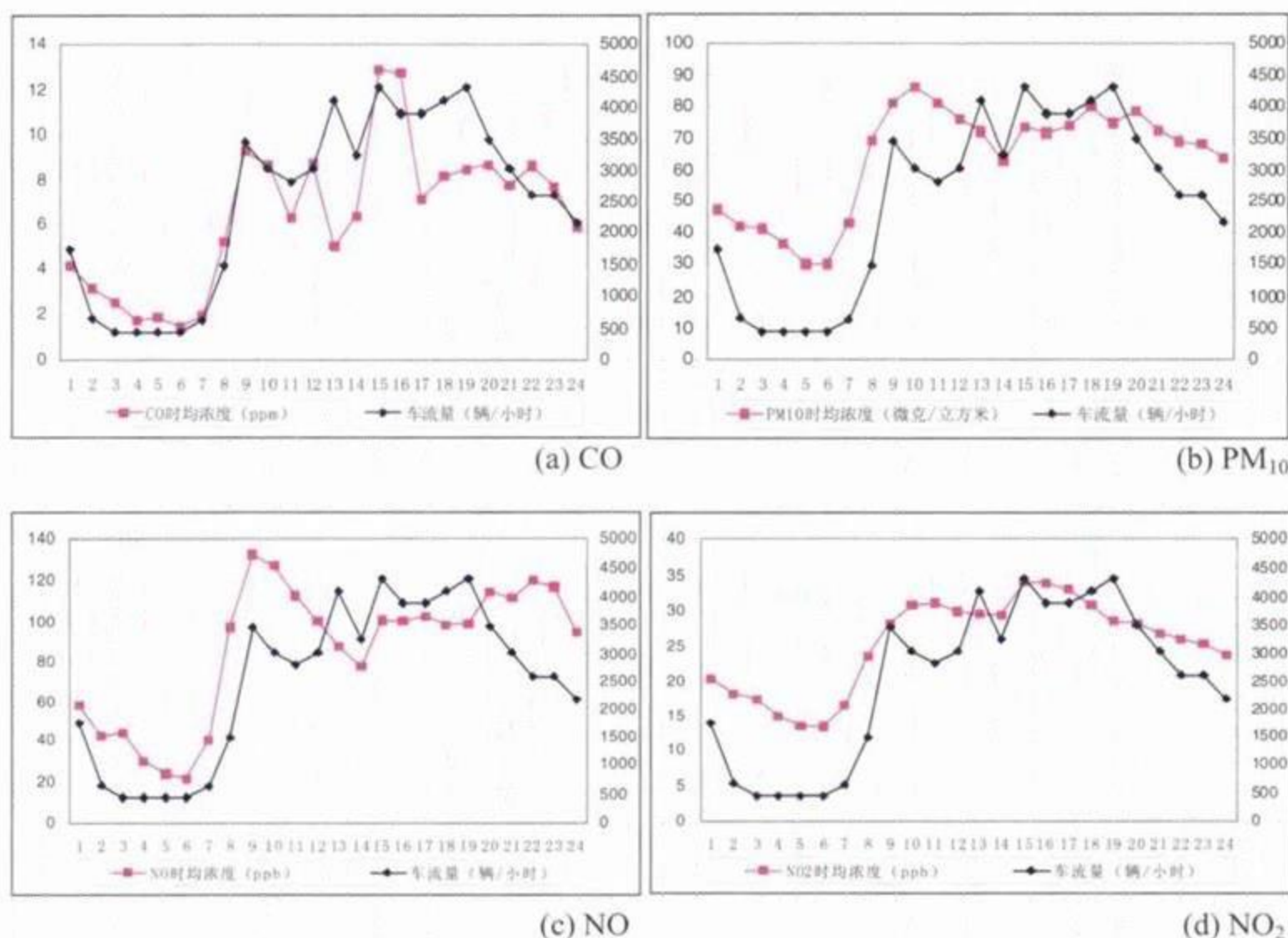


图 6 澳门典型道路边大气污染物浓度与车流量相关性分析

从图中可以得出以下结论：

监测点处于典型道路下风向时，CO、 PM_{10} 、NO 和 NO_2 四种污染物监测浓度的时均值和

车流量时变化都呈现良好的相关性,线性回归得到的 R^2 值在 0.6-0.9 之间,说明该地点的空气质量 and 交通状况密切相关。

根据调研的数据,澳门城区主要街道的车流量中约 20% 是柴油车,因此机动车直接排放的颗粒物对当地大气中的颗粒物浓度有一定的贡献率。

大气中的 NO_2 除了机动车的直接排放外,更主要的是机动车排放的 NO 在大气中被氧化的产物。对水坑尾、大潭山和化验所的三个监测点进行一年监测数据的统计,得到 NO_2 和 NO 的平均浓度比值分别为: 0.68、2.25 和 10.77。而对上述的水坑尾街监测点 36 天的数据做统计, NO_2 和 NO 的平均浓度比值为 0.30。由于 NO 在大气中被氧化为 NO_2 需要一定时间,上述数据表明路边环境的 NO_x 中尚有很大一部分 NO 未转化成 NO_2 。

5. 结论

对于澳门地区的汽油轿车,无论是双怠速检测还是实际运行工况下的随车监测,化油器车的 CO 、 HC 和 NO_x 排放都明显高于安装电喷加三元催化设备的新车。在实际运行循环的启动阶段,由于油温较低,此时催化设备容易出现失效。二冲程摩托车的 HC 排放水平远大于四冲程摩托车,因此淘汰前者,以四冲程摩托车来替代的措施将能从很大程度上降低摩托车的总 HC 排放。此外,根据澳门三个空气质量自动监测站的数据分析,发现交通干道边的 NO_x 浓度比居民区和背景点高一倍以上, PM_{10} 浓度也偏高。交通干道水坑尾街的车流量时变化和监测得到的路边 CO 、 PM_{10} 、 NO 和 NO_2 时均浓度间呈现良好的相关性,说明该地点的空气质量 and 交通状况密切相关。

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Study on Characteristics and Effects of Vehicular Tailpipe Emissions in Macao

Hao Jiming, Hu Jingnan, Wu Ye, Fu Lixin

(Department of Environmental Science and Engineering, Tsinghua University)

Wang Zhishi, Tang Uwa

(School of Science and Technology, Macao University)

Abstract: Vehicular tailpipe emissions are the dominant source to air pollution in Macao and have an important effect on ambient air quality, so there is the necessity to study their characteristics and effects. Two speed idle tests in this article were conducted in the Vehicle Inspection Station of Macao in twice during 2000-2001, including 37 samples of gasoline cars and light-duty vehicles and 20 samples of motorcycles. During the same period, on-board monitoring of tailpipe emissions of another seven gasoline cars were carried out at the real-world status with AVL five-gas analyzer. The results indicate that carburetor vehicles (most are old cars registered before 1995) have much higher emissions of CO, HC and NO_x than newer ones with EFI and TWC within the gasoline cars, either at two speed idle or real world status. Under idle tests CO average concentrations by volume within their emissions are 2.73% and 0.01%, HC 360ppm and 54ppm (hexane equivalence), respectively. Under on-board monitoring NO_x average concentration by volume in emissions from carburetor vehicles is 943ppm, while that of the latter is 140ppm. Analytic converters have low efficiency during the start period of driving routes because of the low oil temperature. There is a significant difference between HC emissions from 2-stroke and 4-stroke motorcycles. HC average concentration by volume in emissions from the former is 5,957ppm (hexane equivalence) under idle tests, which is much larger than that from 4-stroke motorcycles of 238ppm. Besides, during the effect analysis of vehicular emissions in Macao, it's found that NO_x concentration on the side of an arterial road is more than double of that in residential area or at background site and PM₁₀ is also relatively higher by comparing the data of three automatic monitoring stations. Correlation analysis is conducted between the hourly variation of traffic volumes on this road and that of measured roadside pollutant concentrations. It appears to be a good correlative relationship between traffic volumes and concentrations of PM₁₀, NO₂, NO and CO, with the value of R^2 varies from 0.6-0.9 by linear regression.

Keywords: tailpipe emissions; two speed idle tests; real world status; correlation

遥感技术支持下的澳门土地利用解译

胡远安, 程声通, 王建平

(清华大学环境科学与工程系)

王志石, 邓宇华

(澳门大学科技学院)

摘要: 遥感技术可以有效的监测与分析城市土地利用状况。对澳门地区 1988、1992、1995、1997 年 TM 图像进行分析, 可以清晰的观测到这一时期澳门的城市扩张和土地利用变化: 澳门土地通过泥沙沉积和人工填海迅速扩张, 城市扩张主要集中在氹仔和路环。

关键词: 遥感, 土地利用, 澳门

遥感是在不直接接触测量对象的情况下, 对目标物或自然现象远距离感知的一种探测技术^[1]。由于能够大范围高频率的获取地面信息, 遥感数据已经成为城市研究的重要数据源。目前, TM 图像用于土地利用调查已经有较为成熟的经验, 实践证明: 利用 TM 影像可以在小于 0.02 平方公里的区域内划分 18-20 种土地利用类型, 所有一级土地利用分类可以达到 90% 的精度, 分类量算精度可以达到 85% 以上^[2, 3]。

1. 研究背景

澳门是世界上人口密度最大的地区之一。其土地利用方式具有以下特点: 区域面积小, 用地结构复杂, 建筑群集中等。随着澳门城市化的继续推进, 城市环境的评价、预测以及城市土地利用现状、城市变化等问题必然成为城市规划与决策者所面临的核心问题。尤其是人口的巨大压力, 使得澳门的土地利用问题显得尤其突出。

在本次研究中, 主要数据源是四景 TM 影像, 其成像日期分别为: 1988 年 11 月 24 日、1992 年 1 月 20 日、1995 年 12 月 30 日、1997 年 11 月 1 日。所有图像均包括 7 个波段的数据。成像时天气状况良好, 在研究区域上空无云彩, 对图像分析十分有利。图像处理使用的主要软件为 PCI。PCI 是由多个软件包组成的遥感图像处理专用软件, 可以完成包括辐射度校正、几何校正、图像增强、特征提取、图像分类等处理工作, 并支持地理信息系统等多种信息的集成。

2. 分类前数字图像处理

在利用遥感图像进行土地利用解译前, 必须进行预处理, 主要内容如下:

2.1. 图像切割

由于所购影像的区域范围远远大于研究区域, 在进行图像处理前, 首先生成了包含研究区域的子图, 再进一步提取澳门的陆地与水域。

由于缺少澳门边界图, 在本次研究中, 采取了直接在遥感影像上划分边界的方法:

- 1、去除澳门以外的陆地信息;
- 2、利用 TM5 波段确认海陆边界。TM5 波段具有以下特征: 水面的灰度值远远小于陆地, 因此利用阈值法即可区分水体与陆地。

以上均通过编程实现。必须指出的是, 在本次研究中所指的土地面积包括了澳门所有的陆地面积、非海域水面、填海及泥沙淤积面积, 有别于澳门的统计数据。

2.2. 几何校正

要使图像正确的反映地理信息，必须经过几何校正。另外，由于具有统一的地理坐标，上述四幅成像于不同时期的遥感图像可用于叠加分析，更加直观的分析澳门的城市演变过程。在本次研究中，是利用地面控制点进行几何校正，并建立所需的地理坐标。地面控制点主要依据其在图像上的可识别性进行选择，一般选择在桥头、路口等处。

由于澳门地势较为平坦，纠正变换方法采用多项式纠正法，重采样方法采用最邻近单元抽样法。上述方法已经能够获得满意的结果。校正误差均控制在 0.5 个像素(约 15 米)以内。

以 1997 年的几何校正结果为例：

表 1 几何校正误差

测点	M2	M3	T1	C2	C3	C4	C5
误差	0.48	0.49	0.47	0.24	0.12	0.36	0.42

几何校正后的图像地面分辨率可达 25 米。

2.3. 图像特征提取

为了有效的进行图像判读及分析处理，需要从图像数据中求出有益于分析的判读标志及统计量等各种参数，把图像所具有的性质进行定量化的处理，这一过程即为图像特征提取。在本次研究中所采用的特征提取方法主要为光谱特征提取，就是对原始的多光谱进行一定的转化形成一组能够更好的描述地物光谱特性的模式，主要尝试了以下三种方法：

主成分分析：将原始波段的特征轴进行旋转变换，从而得到新的特征轴，使得少数的特征轴可以包含绝大部分的信息；在本次研究中，以 1988 年为例，各主成分轴包含的信息量如下：

表 2 主成分分析信息量

年份	主成分 1 (%)	主成分 2 (%)	主成分 3 (%)
1988	84.18	13.01	1.99

穗帽变换：对除 TM6 之外的 6 个波段进行转化，得到三个分量，分别代表亮度、绿色、湿度，即土壤、植被和水份信息。

比值变换：包括简单比值变换（TM4/TM3,TM3/TM2.TM2/TM1 等）和生物量比值变换（NDVI 等），前者可以有效的去除遥感图像地形起伏的影响，而后者在植被识别中有重要应用^[6, 11]。

3. 土地利用分类

根据土地利用分类规程，一级土地利用类型包括：耕地、园地、林地、牧草地、居民点及工矿用地、交通用地、水域和未利用地。在图像识别中，获取的信息是土地覆盖信息，与土地利用信息仍然有一定的差异。为了保证遥感分类的易辨性和稳定性，在本次研究中，将澳门土地利用类型分成四大类：居民点与工矿用地、交通用地和未利用地、水域、绿地(包括林地与草地)。

在本次研究中，土地利用分类所使用的方法是监督分类。

3.1.1. 训练区光谱

监督分类主要是依据遥感图像上已知类别属性的像元，分析出该类别地物的光谱特征，并据此对其它地物进行分类。为了减少分类的人为误差，对不同成像日期所取的训练区尽量

统一。训练区的原始波段光谱特征见下表（以 1988 年的图像为例）：

表 3 原始波段光谱信息

土地利用类型	TM1	TM2	TM3	TM4	TM5	TM6	TM7
居民点、工矿用地	85.4	36.9	39.7	31	43.2	113.9	27.3
交通、未利用地	96.2	47.9	58.7	56.2	102.9	114.5	54.6
绿地	74	30.5	29.6	41	41.2	111.3	16.3
水域	78.8	33.3	31.1	18.6	11.1	110.3	5.8

由上表可知，不同地物在不同波段的可分离性存在着较大的差异。
提取后的特征如下（以 1988 年的图像为例）：

表 4 特征波段信息

土地利用类型	主成分			缨帽分析			比值变换		
	1	2	3	1	2	3	ID	T ₄₃	T ₄₅
居民点、工矿用地	150.1	138.7	117.7	41.7	89.2	182.1	114.7	78.6	65.2
交通、未利用地	210.7	170.7	128.1	97.4	80.6	147.5	12.3	88.0	54.1
绿地	150.4	112.5	127.2	36.4	157.7	191.0	150.2	164.4	110.7
水域	102.6	123.7	119.9	7.8	79.2	217.9	94.1	50.8	182.1

注：ID=（TM4-TM3）/（TM4+TM3）×100+127；T₄₃=TM4/ TM3×100；T₄₅=TM4/ TM5×10

由上表可知：主成分分析对数据具有显著的压缩作用，从第一主成分到第三主成分，信息量减少趋势非常明显；缨帽分析和比值变换对于放大某一特定地物与其它地物的差别具有较好的作用。

3.1.2. 试分类

在本次研究中，主要尝试了四组分类：

利用不同数量的波段组合对土地进行分类：以 1997 年图像为例，使用的波段组合包括 TM4，TM4-5,TM3-4-5,YM3-4-5-7,TM1-3-4-5-7,TM1-2-3-4-5-7；

利用相同数量的不同波段组合对土地利用进行分类：对所有四景图像利用下列组合进行分类：TM1-4-5,YM2-4-5,YM3-4-5,根据统计信息由 PCI 选择的最佳波段；

利用主成分分析和缨帽分析后的结果进行分类：主成分 1-2-3,主成分 1-2,缨帽分析 1-2-3；
利用比值变换后的结果进行分类。

3.1.3. 分类结果比较

对以上用不同方法获得的分类结果进行比较，主要判据如下：

“训练区”样本的可分离度：波段能否有效分离地物，可以用 Bhattacharrya（0-2）距离表示，0 为完全不可分离，0—1 为不可分离，1—1.9 为可分离性较差，1.9 以上可分离性好；

“训练区”样本的混淆矩阵，混淆矩阵是对“训练区”内各像元分类后的信息统计，对角线元素越接近 1，精度越高；

分类图像对比判读：对用不同分类方法获取的结果进行叠加，分析分类的主要差异及差异产生的可能原因。

比较结果简述如下：

TM3-4-5,YM3-4-5-7,TM1-3-4-5-7,TM1-2-3-4-5-7 的样本可分离度和混淆矩阵均能够满足要求,而且分类结果十分接近,有差异的像素量小于 10%;

主成分 1-2-3 的分类结果与上述原始波段组合接近,主成分 1-2、缨帽分析 1-2-3 的分类结果不理想;

比值变换的除绿地外,对其它地物产生了明显的误分类。

综上所述,可以使用三波段以上的组合及主成分 1-2-3 分类的成果进行进一步处理与分析。

3.2. 分类结果修正

以上通过单一方法分类产生的结果往往包含一定的误差。但大多数误差可以根据具体情况分析去除,以进一步提高分类精度。

以下是对部分误分类情况的分析以及解决方法:

表 5 误分类及其修正

误分类区域	主要现象	原因	解决方法
水陆边界像元	相当数量的水陆边界附近的像元被划为居民点和工矿用地	水体与陆地综合作用的光谱特征与居民点、工矿用地的光谱特征非常接近。	沿水体生成 buffer 区,并去除误分类的像素
居民点及工矿用地	澳门半岛部分居民点和工矿用地被划分为水域	岛内建筑物密集,受阴影等因素的影响显著。	利用滤波删除零碎图斑。
桥梁	大桥被划分为水域	桥梁光谱信息被水体光谱信息所掩盖。	根据已知情况修正。

修正后的分类结果见下表:

表 6 澳门土地利用

年份	1988	1992	1995	1997
居民点及工矿用地 (km ²)	3.53	4.45	5.44	5.71
交通用地及未利用地 (km ²)	4.88	6.95	7.61	7.16
绿地 (km ²)	8.74	6.79	8.84	9.93
水域 (km ²)	0.55	0.52	1.32	1.36

4. 澳门土地利用结果分析

澳门的土地面积持续增长,在 92—96 年间增长速度最快。不同岛屿在不同时间段的增长速度如下:

表 7 澳门土地扩张

岛屿	88/11/24—92/1/20	92/1/20—95/12/30	95/12/30-97/11/1
澳门 (km2/年)	0.03	0.47	0.01
氹仔 (km2/年)	0.21	0.57	0.31
路环 (km2/年)	0.09	0.12	0.19

居民点与工矿用地不断增长,但在近期有放缓的趋势。下表是澳门的居民点用地扩张情况:

表 8 居民点与工矿用地增长

岛屿	88/11/24—92/1/20	92/1/20—95/12/30	95/12/30-97/11/1
澳门 (km2/年)	0.19	0.09	0.04
氹仔 (km2/年)	0.07	0.11	0.08
路环 (km2/年)	0.01	0.06	0.02

从土地利用分布的情况来看,澳门半岛始终是居民点和工矿用地的集中地,但是目前已经接近饱和,而氹仔的增幅较大,而且这种趋势仍有继续的可能;氹仔的可利用的未利用地在三个岛屿中最多,具有最大的发展可能;路环的植被保持状况良好,居民点和工矿用地的增长相对缓慢。

从卫星图像上来看,澳门半岛附近几乎没有泥沙沉积现象,路氹公路之间及以东、以西区域是泥沙的主要沉积区域,是填海的理想地,路环以南部分区域泥沙沉积较少,可以建设新港。

5. 结语

综上所述,利用 TM 图像能够快速有效的进行澳门城市演变的动态观测,但受地面分辨率的限制较大。建议在今后的研究中采用更高精度的卫星影像或航空图像,以进一步提高对城市用地的分类精度。

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Analysis of macau land use with the tool of remote sensing

Hu Yuanan, Cheng Shengtong, Wang Jianping

(Environmental Science and Engineering Department, Tsinghua University)

Wang Zhishi, Deng Yuhua

(College of Science and Technology, Macau University)

Abstract: Remote sensing is an effective tool to monitor and analyze the land use of urban area. The urban expansion and land use change of macau, which can be observed definitely from 4 TM images of different years(1988,1992,1995,1997), has characteristics as following: the land area expanded by the way of sediment deposit and sea reclamation; the urban expansion orients to taipa and coloane.

Keywords: remote sensing; land use; macau

澳门海域的水色遥感反演研究*

况昶, 王建平, 程声通, 贾海峰

(清华大学环境科学与工程系, 北京 100084)

王志石, 邓宇华

(澳门大学科技学院, 澳门)

摘要: 本文以澳门海域的水色遥感反演为案例扼要地介绍了水色遥感反演的全过程。在同步监测的基础上, 获取遥感数据, 然后对遥感数据进行了预处理, 包括几何校正、辐射校正和大气校正, 最后拟定不同反演方案, 进行相关性分析。在笔者进行大量模拟计算的基础上, 得出一些非常有意义的结论: 由于悬沙浓度的影响, 采用分区处理的反演技术能使自变量和因变量的相关性有明显提高, 同时分析了不同大气校正方案的效果。

关键字: 分区处理 水色遥感 反演 澳门海域

澳门沿海是典型的第二类水体, 悬砂浓度高, 对水体光谱信息的影响最大。叶绿素和黄色物质也对水体光谱信息有一定的影响。澳门位于珠江河口西部, 西江河口东部, 与珠海经济特区毗邻, 三面环海。由于该地区河流众多, 且多为径流河口, 每年将大量泥沙带入海中, 导致该地区海岸水独特的水文特性: 高浊度、海岸线活动剧烈、潮汐弱、水域狭窄且浅等。这些特点不利于城市污水排放的扩散, 造成澳门周围水域水质持续恶化。

对沿海、河口或内陆水体 (Case 2, 第二类水体) 进行水色遥感, 通过反演叶绿素、悬砂以及黄色物质的浓度和分布, 对水体污染、赤潮监测、富营养化调查有一定的指导作用。水色遥感具有大范围、同步性和相对成本较低等优点, 能有效地提高环境监测的时间频率和空间覆盖面。特别地, 叶绿素浓度是沿海地带或大型湖泊、水库的水质的重要指标, 是水体营养化程度的重要标志。它能够反映水体中的营养物质的含量及分布, 并间接反映水体的有机污染程度。

1. 数据

1.1. 水质数据

水质数据的采样于 1997 年 11 月在澳门沿海进行, 采样点为 25 个, 如图 1 所示。图中的 A 为澳门半岛, B 为氹仔岛, C 为路环岛。水样点如右图表示, 浅色点表示近区, 特点是水较浅, 租用小船采样; 深色点表示远区, 特点是水较深但航程远, 租用大船采样。两艘船各配备以下仪器: PH 计 (兼测温度)、盐度计、DO 仪。

每个点均测量了悬砂、叶绿素 *a*、总有机碳、总无机碳、总氮、溶解氧、盐度、温度、PH 等指标。其中, 溶解氧、盐度、温度、PH 是在现场实测, 其余指标在实验室测定。另外,

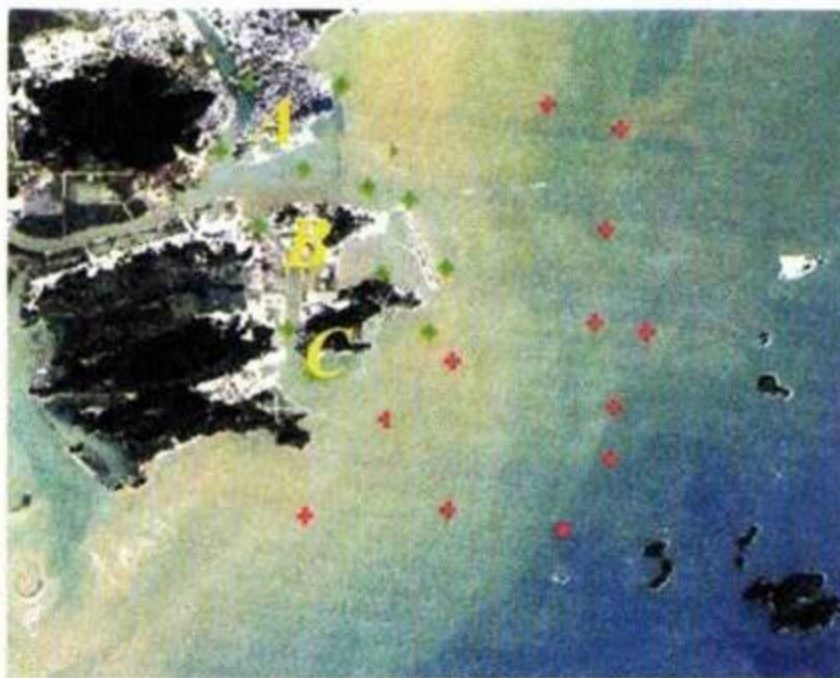


图 1 水样测点分布图

国家自然科学基金委员会资助项目(49971058), 国家教育部博士点基金资助项目(98000368)

澳门基金会与澳门大学资助项目

各点均使用 Garmin GPS 接收仪确定经度和纬度。

1.2. 卫星数据

卫星数据为准同步的 Landsat 5 TM 数据，轨道号为 122/44。图像清晰，无云彩。值得注意的珠江三角洲地区常年多云，深秋至冬季才出现少云天气。

1.3. 遥感数据预处理

在开发模型前对遥感数据进行了预处理，包括几何校正、辐射校正和大气校正。考虑到本部分研究集中在水体，还需要分割陆地和水域。为了后续分析，本文着重介绍一下水陆分割和大气校正两部分。

1.3.1. 水陆分割

考察 TM 的各个波段，发现 4、5、7 三个波段对于水陆分割很有用。图2 是 TM 波段 4、5 的灰度值直方图。横坐标为灰度值，刻度分别为 64、128、192、256；纵坐标为对应灰度值的象元占全部象元数的百分比。可以看出，波段的灰度分布集中在两个区域，中间有一个明显的下凹点。该点对应灰度就是水体、陆地的区分依据。本研究使用的分割判据是 $TM5=10$ 。

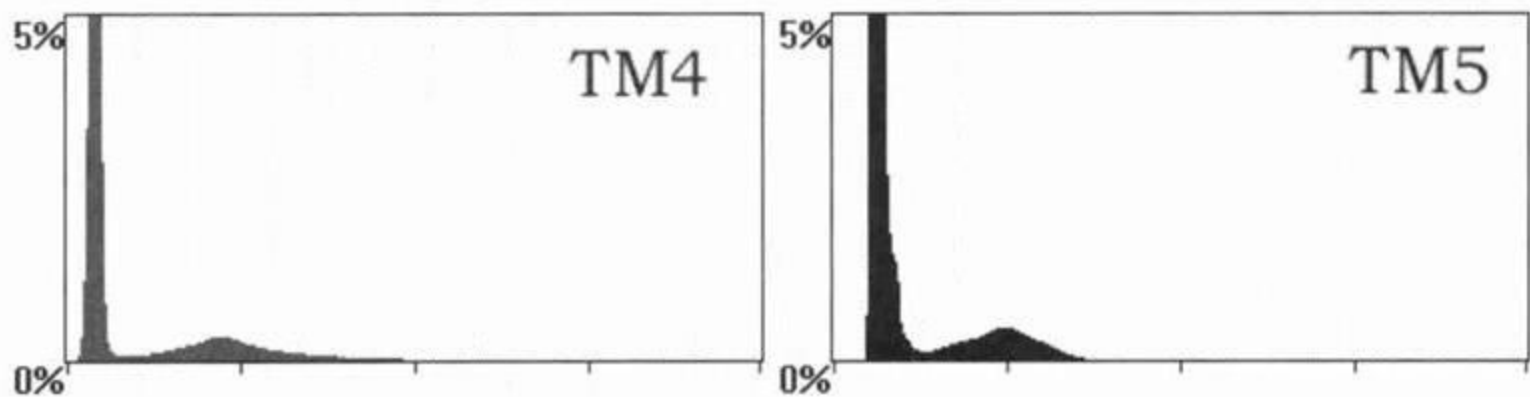


图 2 TM 波段 4、5 的灰度值直方图

1.3.2. 大气校正

PCI 提供了一套针对陆地传感器（TM/MSS/SPOT）的大气校正命令 ATCOR0、ATCOR1 和 ATCOR2，流程如图3 所示。



图 3 PCI 提供的大气校正流程图

此方法的理论依据由里克特（Richter）给出^[iii, iv]，其中心思想是：按照标准大气的分类，计算不同气溶胶类型、太阳天顶角、地面海拔高度、大气能见度下的大气散射，并存放为一

个类似查找表的目录。在实际应用时按照查找表进行相应的大气校正。里克特使用的分类依据来自中分辨率大气透射模型 MODTRAN^[1]。他的算法还考虑了地面反射的邻接效应并进行了校正，最后输出地面辐射率。

2. 反演研究

2.1. 拟合自变量和因变量

拟合自变量 R_i 来自各波段灰度值，但不是简单的灰度值，而是考虑了是否对图像作大气校正、是否作均值滤波（以去除噪声的影响），总共 12 种情况。

- ✓ 大气校正：未作大气校正、“减黑体法”大气校正、ATCOR 系列大气校正。共计 3 种情况。
- ✓ FAV 卷积核：未作均值滤波、 3×3 、 5×5 、 7×7 。共计 4 种情况。

FAV 指均值滤波，研究中对比了不同的卷积核带来的影响。
拟合因变量也考察了两种情况：组份浓度，浓度对数值。

2.2. 方案 1

表 1 列出了 75 种波段组合（文献^[1]），表 2 列出了 81 种波段组合（笔者拟定的组合方案）。

表1 波段组合 1

$R1$	$R2$	$R3$	$R5$
$R2/R1$	$R3/R1$	$R3/R2$	$R3/(R1+R2)$
$(R2+R3)/R1$	$(R2*R3)/R1$	$R5/R1$	$R5/R2$
$R5/R3$	$R5/(R1+R2)$	$R5/(R1+R3)$	$R5/(R2+R3)$
$R5/(R1+R2+R3)$	$(R3+R5)/(R1+R2)$	$(R3+R5)/(R1*R2)$	$(R3*R5)/(R1+R2)$
$(R3*R5)/(R1*R2)$	$(R3+R5)/R1$	$(R3+R5)/R2$	$(R3*R5)/R1$
$(R3*R5)/R2$	$R2+R3$	$R2*R3$	$R3+R5$
$R3*R5$	$\ln(R1)$	$\ln(R2)$	$\ln(R3)$
$\ln(R5)$	$R2/\ln(R1)$	$R3/\ln(R1)$	$R3/\ln(R2)$
$R3/\ln(R1+R2)$	$(R2+R3)/\ln(R1)$	$(R2*R3)/\ln(R1)$	$R5/\ln(R1)$
$R5/\ln(R2)$	$R5/\ln(R3)$	$R5/\ln(R1+R2)$	$R5/\ln(R1+R3)$
$R5/\ln(R2+R3)$	$R5/\ln(R1+R2+R3)$	$(R3+R5)/\ln(R1+R2)$	$(R3+R5)/\ln(R1*R2)$
$(R3*R5)/\ln(R1+R2)$	$(R3*R5)/\ln(R1*R2)$	$(R3+R5)/\ln(R1)$	$(R3+R5)/\ln(R2)$
$(R3*R5)/\ln(R1)$	$(R3*R5)/\ln(R2)$	$\ln(R2+R3)$	$\ln(R2*R3)$
$\ln(R3+R5)$	$\ln(R3*R5)$	$\ln(R3*R5)/\ln(R1+R2)$	$\ln(R3*R5)/\ln(R1*R2)$
$\ln(R3*R5)/\ln(R1)$	$\ln(R3*R5)/\ln(R2)$	$\ln(R3+R5)/\ln(R1+R2)$	$\ln(R3+R5)/\ln(R1*R2)$
$\ln(R3+R5)/\ln(R1)$	$\ln(R3+R5)/\ln(R2)$	$R3-R5$	$(R3-R5)/R1$
$(R3-R5)/R2$	$(R3-R5)/\ln(R1)$	$(R3-R5)/\ln(R2)$	$\ln(R3-R5)/\ln(R1)$
$\ln(R3-R5)/\ln(R2)$	$\ln(R3-R5)/R1$	$\ln(R3-R5)/R2$	

表 2 波段组合 2

$\ln(R1)/R2$	$R1/\ln(R2)$	$R1/R2$	$\ln(R1/R2)$
$\ln(R1/R3)$	$\ln(R1)/R3$	$\ln(R1)/\ln(R2)$	$R1/R3$
$R2/R3$	$\ln(R2/R3)$	$R1/\ln(R3)$	$\ln(R1)/\ln(R3)$
$\ln(R2)/\ln(R3)$	$R1/(R2+R3)$	$\ln(R2)/R3$	$R2/\ln(R3)$
$\ln(R1)/\ln(R2+R3)$	$\ln(R1)/(\ln(R2)+R3)$	$\ln(R1)/(R2+R3)$	$\ln(R1)/(R2+R3)$
$R1/\ln(R2+R3)$	$R1/(\ln(R2)+R3)$	$\ln(R1)/(R2+\ln(R3))$	$\ln(R1)/(\ln(R2)+\ln(R3))$
$R1/(R2*R3)$	$\ln(R1/(R2*R3))$	$R1/(R2+\ln(R3))$	$R1/(\ln(R2)+\ln(R3))$
$\ln(R1)/(\ln(R2)*R3)$	$\ln(R1)/(R3*\ln(R3))$	$\ln(R1)/(R2*R3)$	$\ln(R1)/\ln(R2*R3)$
$R1/(\ln(R2)*R3)$	$R1/(R2*\ln(R3))$	$\ln(R1)/(\ln(R2)*\ln(R3))$	$R1/\ln(R2*R3)$
$\ln((R1+R2)/R3)$	$\ln(R1+R2)/R3$	$R1/(\ln(R2)*\ln(R3))$	$(R1+R2)/R3$
$(\ln(R1)+\ln(R2))/R3$	$(R1+R2)/\ln(R3)$	$(\ln(R1)+R2)/R3$	$(R1+\ln(R2))/R3$
$(R1+\ln(R2))/\ln(R3)$	$(\ln(R1)+\ln(R2))/\ln(R3)$	$\ln(R1+R2)/\ln(R3)$	$(\ln(R1)+R2)/\ln(R3)$
$\ln(R1*R2)/R3$	$(\ln(R1)*R2)/R3$	$(R1*R2)/R3$	$\ln((R1*R2)/R3)$
$(R1*R2)/\ln(R3)$	$\ln(R1*R2)/\ln(R3)$	$(R1*\ln(R2))/R3$	$(\ln(R1)*\ln(R2))/R3$

$(\ln(R1)*\ln(R2))/\ln(R3)$	$(R1+R3)/R2$	$(\ln(R1)*R2)/\ln(R3)$	$(R1*\ln(R2))/\ln(R3)$
$(\ln(R1)+R3)/R2$	$(R1+\ln(R3))/R2$	$\ln((R1+R3)/R2)$	$\ln(R1+R3)/R2$
$\ln(R1+R3)/\ln(R2)$	$(\ln(R1)+R3)/\ln(R2)$	$(\ln(R1)+\ln(R3))/R2$	$(R1+R3)/\ln(R2)$
$(R1*R3)/R2$	$\ln((R1*R3)/R2)$	$(R1+\ln(R3))/\ln(R2)$	$(\ln(R1)+\ln(R3))/\ln(R2)$
$(R1*\ln(R3))/R2$	$(\ln(R1)*\ln(R3))/R2$	$\ln(R1*R3)/R2$	$(\ln(R1)*R3)/R2$
$(\ln(R1)*R3)/\ln(R2)$	$(R1*\ln(R3))/\ln(R2)$	$(R1*R3)/\ln(R2)$	$\ln(R1*R3)/\ln(R2)$
$(\ln(R1)*\ln(R3))/\ln(R2)$			

以上共计算了 3744 种情况。十分遗憾的是，相关系数没有比较突出的。由于数据量大，不可能一一列出结果。以下只列出相关系数相对较高的几种情况。

大气校正	FAV	表达式	叶绿素	相关系数
ATCOR	无	$R1/(R2*\ln(R3))$	浓度	0.422
ATCOR	3×3	$\ln(R1)/(\ln(R2)*\ln(R3))$	浓度对数	0.411
减黑体	3×3	$(R1+R3)/R2$	浓度	0.369

- 分析数据可以得出以下一些结论：
- 使用原浓度和对数值进行拟合的结果没有太大区别，在大部分情况下使用对数值后拟合系数稍有提高。
- ✓ 均值滤波 FAV 对拟合结果帮助不大。3×3 的卷积核优于 5×5 和 7×7。
 - ✓ 大气校正中的“减黑体”法和不作大气校正的效果基本一样，ATCOR 大气校正后效果有一定改善。
 - ✓ 似乎没有占优势的波段组合，当然单波段或 2 波段组合的结果都不如 3 波段组合。

以上 3744 种都是一元线性回归。由于效果不好，又作了多元线性回归，即以下 4 种情况。 R_i 仍然是 12 种情况，拟合因变量是 2 种情况。

$R1+R2+R3+R4+R5+R7$	$\ln(R1)+\ln(R2)+\ln(R3)+\ln(R4)+\ln(R5)+\ln(R7)$
$R1+R2+R3+R4$	$\ln(R1)+\ln(R2)+\ln(R3)+\ln(R4)$

所有 3840 种情况的相关系数不超过 0.5。这说明对波段进行花样众多的组合（多元线性回归其实也是波段之间的组合！）不一定能得到满意的结果。

2.3. 方案 2

观察测点分布和数据规律。小船测点距离海岸近，水域的浊度高，悬砂浓度高，不妨称为近区。大船测点距离海岸远，水域的浊度相对低，悬砂浓度更低，可以称为远区。表3 给出了两个区域的悬砂和叶绿素浓度均值，近区的悬砂浓度明显高于远区（3.4 倍），但叶绿素浓度在同一数量级。

表 3 近区和远区的悬砂/叶绿素浓度均值

	点数	悬砂浓度均值 (mg/L)	叶绿素浓度均值 (mg/m ³)
近区	13	42.04	0.870
远区	12	12.21	0.716

这启发我们提出一个猜想：两个区域不仅仅是与海岸的物理距离不同，水体组份的分布也是不同的，应该分别处理。鉴于波段组合不一定能得到满意的结果，使用了多元线性回归的方法。表4 简单给出了部分有代表性的结果。第 1 列的表达式中，“-”前面的是拟合因变量，有两个拟合因变量的情况是指它们一起参与拟合。“-”后面的是拟合自变量， $R16$ 表示波段 1 到 6 的灰度值， $\ln(R16)$ 为灰度值的对数。

表 4 各种情况下远区和近区的拟合结果

区域 →	远区				近区			
大气校正 →	ATCOR		无大气校正		ATCOR		无大气校正	
均值滤波 →	无 FAV	3×3	无 FAV	3×3	无 FAV	3×3	无 FAV	3×3
$C-R16$	0.896	0.830	0.873	0.912	0.707	0.615	0.553	0.743
$LnC-R16$	0.781	0.640	0.785	0.372	0.618	0.733	0.633	0.764
$C-ln(R16)$	0.900	0.872	0.887	0.938	0.759	0.628	0.615	0.854
$LnC-ln(R16)$	0.894	0.771	0.859	0.875	0.714	0.571	0.559	0.754
$S-R16$	0.847	0.663	0.824	0.426	0.772	0.852	0.775	0.857
$LnS-R16$	0.905	0.855	0.894	0.912	0.756	0.579	0.593	0.784
$S-ln(R16)$	0.859	0.806	0.851	0.904	0.838	0.688	0.723	0.724
$LnS-ln(R16)$	0.725	0.662	0.793	0.387	0.681	0.696	0.583	0.735
$C, S-R16$	0.894	0.857	0.869	0.938	0.838	0.705	0.736	0.862
$lnC, lnS-R16$	0.857	0.745	0.842	0.868	0.811	0.663	0.752	0.751
$C, S-ln(R16)$	0.779	0.667	0.816	0.399	0.643	0.814	0.719	0.836
$lnC, lnS-ln(R16)$	0.881	0.828	0.884	0.914	0.813	0.665	0.752	0.799

可以看出：

- ✓ 分区后两个区域的线性拟合效果明显要优于放在一起拟合的效果，充分说明两个区域水体组份的物理性质有所不同。
- ✓ 对叶绿素和悬砂，远区的效果均好于近区，也说明了近区由于悬砂浓度过高而带来的干扰更大。
- ✓ 通过 ATCOR 大气校正后再进行拟合的结果要优于不作大气校正，说明比较准确的大气校正还是很有必要的。
- ✓ 一般来说，不作 FAV 的效果比 FAV (3×3) 要好，特别是对远区而言。
- ✓ 叶绿素取对数时的效果好于不取对数的效果，但悬砂不取对数时的效果更好。灰度值取对数时的效果要优于不取对数时的效果。

特别要注意第 5 点，在实际应用时叶绿素和悬砂的浓度还是应该取对数的。虽然使用不取对数时的拟合结果来反演组份浓度对自变量不敏感，但可能会出现负数！

3. 结论与讨论

3.1. 分区处理技术

由以上分析可知，分区后两个区域的线性拟合效果明显要优于放在一起拟合的效果，充分说明两个区域水体组份的物理性质有所不同，在进行水色遥感反演时需要分别处理。同时从前边的分析表明，ATCOR 大气校正后再进行拟合的结果要优于不作大气校正，说明比较准确的大气校正对统计反演模型也还是很有必要的。一般来说，不作 FAV 的效果比 FAV (3×3) 要好，特别是对远区而言。

3.2. 问题讨论

分区处理的需要解决两个问题：①如何划分区域？显然，近区和远区要包含各自相应的测点，两个区域不能相交。这样，两个区域之外的水域姑且称为过渡区。②过渡区的叶绿素浓度如何反演呢？当然不能使用近区公式或远区公式。比较科学的方法是利用两个区域的公式进行反演后再加权平均。

分区方案如图 4 所示, 基本是沿着(目视)可见的悬砂泥沙的走势划分的。计算过渡区浓度的权重系数如下选取: 先求解所在点与近区的最短距离 dis_n 和与远区的最短距离 dis_f , 则近区对应的权重系数是 $dis_f / (dis_f + dis_n)$, 远区对应的权重系数为 $dis_n / (dis_f + dis_n)$ 。这种确定权重的方法不见得有多少理论依据, 但处理起来比较简单, 因为不涉及到水体本身的性质。



图 4 近区和远区的范围

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The Inversing Study of Water Color Remote Sensing in Sea Area of Macau

Kuang Chang, Wang Jianping, Cheng Shengtong and Jia Haifeng

(Dept. of Environmental Science and Engineering, Tsinghua University, Beijing 100084, PRC)

Wang Zhishi and Tang U Wa

(University of Macau, Faculty of Science and Technology, Macau, PRC)

Abstract: Selecting Macau sea area as a case, a full inversing process of water color remote sensing was presented. In the research, firstly a TM synchronous monitoring was taken and the TM image data was acquired, then the remote sensing data was pretreated, these included geometric correction, radiation correction and atmospheric correction. Finally different inversing schemes were proposed and analyzed. After large simulating calculation, a significant conclusion was found: because of the influence of SS concentration, if the technology of subarea inversing was adopted, the correlation of dependent and independent variables would be improved greatly. In this paper the inversing effect of different atmospheric correctiongs were also analyzed.

Keywords: subarea processing; Water color remote sensing; inversing; Sea area of Macau

小型湖泊生态系统模型及其不确定性分析 ——以澳门南湾湖水质模拟为例

曾思育, 杜鹏飞, 冉圣宏, 陈吉宁

(清华大学环境系统分析研究所, 北京, 100084)

王志石, 邓宇华

(澳门大学科技学院)

摘要: 本文以澳门南湾湖为例, 对一个小型湖泊生态系统模型的基本结构功能进行了描述。并且在进行水质模拟过程中, 利用两种不同的不确定性分析算法对参数进行率定, 讨论了参数不确定性, 以及传递到计算结果中对水质模拟的不确定性影响; 在此基础上, 对模型输入(污染负荷)的不确定性也进行了粗略研究。

关键词: 湖泊模型, 不确定性分析, GLUE 算法, HSY 算法

1. 引言

澳门南湾湖是人工开挖而成的小型湖泊, 既没有上游来水和支流汇入, 又没有常规的下游出流; 因此与外界环境中物质能量的交换比较少, 可以说整个湖泊构成了一个较为封闭的生态系统。针对这样的小型湖泊, 以藻类的生长死亡为核心, 模拟湖泊生态系统所涉及的生物化学过程, 从而预测湖泊的水质变化, 是建立湖泊生态系统模型的任务。由于南湾湖的形成时间短, 相关的监测资料和监控数据不多, 尤其是有关污染物来源和污染负荷的信息量比较少, 在运用湖泊模型模拟水质的动态变化时, 无疑存在很大的不确定性, 例如模型参数的不确定性和污染物总量输入上的不确定性, 为此, 对模型不确定性的分析也成为本文讨论的内容之一。

2. 模型基本功能与结构

笔者针对澳门南湾湖开发了小型湖泊生态系统模型。对湖泊水体本身而言, 这是一个单箱动态模型, 但同时考虑了底泥对湖泊水质的影响。具体来说, 模型的功能可以分成三个部分, 即 (1) 水文过程模拟, (2) 热动力学过程模拟, (3) 生物过程及水质模拟。

2.1. 水文过程模拟

对南湾湖而言, 它是一个小型的人造湖体, 其环境功能主要是作为景观娱乐, 对湖泊基本不存在人工调度, 与外界没有频繁的水量交换, 由于地理位置的关系, 湖泊冬季不会出现结冰现象, 因此影响它水位变化的主要是湖面降雨和蒸发这两个因素。因此, 水文模拟子模型的主要任务就是, 描述南湾湖水体体积和水位随着湖面降雨量和蒸发量变化的动态变化过程。在模拟过程中, 每日降雨量是作为已知条件输入的, 而湖面的每日蒸发量则是根据日平均风速、日平均气温和日平均露点等已知的气象数据计算出来的。

2.2. 热动力过程模拟

在这一部分, 模型主要是模拟计算湖泊水温的动态变化, 而不必考虑冬季结冰对湖内生态系统的影响。之所以要模拟计算湖水温度, 是因为水温的变化直接会影响到湖泊生态系统

的状态以及污染物的降解速率。另外,由于南湾湖水深较浅,在模型概化时可以把湖体看成完全混合的单箱,而不必考虑因水温在竖向分布上的实际差异引起的水体分层现象。根据以上特点,南湾湖的热动力学子模型主要描述了由于湖体中水量变化引起的热交换、湖泊表面与大气之间的热交换以及因日照引起的热交换等,包括长波辐射方程、短波辐射方程、气—水对流传热方程、以及蒸发和降雨引起的热量传递。通过热量的动态平衡变化,可以推算出水温的动态变化,进而计算出当前的湖泊温度。

2.3. 生物过程及水质模拟

根据南湾湖现有的监测数据以及未来管理的需要,以藻类新陈代谢过程为主线构建的南湾湖水质模型(或生态系统模型)包括了以下水质项目:藻类、慢速碳(Slow Carbon,相当于COD和BOD的差值)、快速碳(Fast Carbon,相当于BOD)、COD、有机氮、氨氮、硝酸盐氮(包括亚硝酸盐氮部分)、总氮、总磷、溶解氧、大肠杆菌和挥发酚。在这12个水质项目之间存在这错综复杂的关系,尤其是再加上底泥与水体之间的相互作用。湖体内发生的主要生化反应关系可以用图1来表示。

3. 参数率定及其不确定性分析

3.1. 数值计算方法

由于湖泊系统的水质变化与河流系统相比,一般比较缓和,采用有限差分法求解模型的数值解稳定性容易得到满足,因此在南湾湖的实例中,笔者采用了定步长的有限差分法。在差分格式的选择上,软件设计了欧拉法和龙格库塔法两种,前者精度稍差,但计算时间短,后者反之,可由用户自行选择。

3.2. 参数率定算法

由于整个水质模拟过程比较复杂,因而涉及到众多的参数,参数率定成为该模型构建过程中一个非常重要的环节。针对这种结构复杂的模型,笔者采用了基于不确定性分析(Uncertainty Analysis)技术的参数率定方法,具体地,尝试了GLUE算法和HSY算法。图2表示的就是GLUE算法和HSY算法的基本过程。

由于两种算法的原理不同,对参数不确定性的度量和描述方法不同,因此参数率定的结果及其表达形式不同,最终反映到水质预测结果上也会有所不同。以“藻类生长最大速率”这一参数的率定为例,先验的参数分布为(0.35, 3.5)区间上的均匀分布;用2001年1月至3月期间的湖体藻类监测数据(叶绿素a浓度)作为观测数据,分别用两种方法进行率定,且参数空间的采样次数均设定为10000次。GLUE算法给出的率定结果则是采样时所取的10000次参数值及其对应的似然度,而HSY算法给出的结果是使得计算值与观测值之间的绝对误差在 $\pm 0.0045\text{mg/L}$ 范围内对应的若干个(本次算例中为640个)“可信”的参数值(即纵坐标不为0的参数取值),分别如图3和图4。

3.3. 参数不确定性引起的预测结果不确定性

由于参数取值本身存在着不确定性,水质模拟过程中,参数的不确定性必然会传递到预测结果当中去。对于传统的确定性模拟(Deterministic Simulation)计算来说,水质预测值是一个确定的取值;而在基于不确定性分析的模拟计算中,还必须包含有对水质预测结果的不确定性做出的估算。具体来说,基于HSY算法的模型结果应该是由上下限框定的水质预测的取值范围;而基于GLUE算法的计算成果则是水质预测值的概率分布,若给定相应的置信度(例如90%),也可以获得一个取值范围(置信区间)。利用率定的参数结果,可以“预测”2001年4月至6月期间的水质状况。不同的算法结果如图5、图6和图7。

4. 污染物负荷估算的不确定性对湖泊水质模拟的影响

对南湾湖而言,目前已知的污染源主要是湖面降水带来的污染物。但现有的监测资料和相关研究基本没有涉及到降雨中污染物的负荷量,也没有对连续降雨过程中污染物浓度的时间变化过程做过讨论,因此整个湖泊内部水体水质的模拟计算是建立在很粗略的污染物负荷估算基础上的。由此可以想见,水质模拟结果必然强烈地受到污染物负荷估算中不确定性的影响。为此,笔者开发了相应的计算程序,试图探索污染负荷输入的不确定性在湖泊水质模拟过程中的传递。受条件限制,目前仅讨论降雨中总磷负荷的不确定性问题,并且假定每次降雨的不确定性变化程度是一致的。可以看到,当输入到南湾湖的总磷负荷量在原估算量的 $\pm 50\%$ 范围内随机变化时(其它条件全部为确定性的),水质模拟也相应产生一定的不确定性,图8就是利用HSY算法获得的结果。利用模型对南湾湖的水质进行模拟预测,最终的目的是要为将来的湖泊管理和污染控制提供决策支持和依据。根据前文对污染负荷估算不确定性的讨论可知,要想充分发挥模型的作用,还必须对模型输入的不确定性进行深入研究。对南湾湖而言,有必要开展污染源及其污染负荷的详细研究,尤其是降雨中的污染物时空变迁规律,从而降低模型输入的不确定性,最终提高水质预测结果的可靠性。

5. 小结

本文以澳门南湾湖为例,阐述了一个小型湖泊水质模型的具体运用。这个模型的特点在于它是一个基于不确定性分析框架建立起来的,因此具有不确定性分析的一些基本功能,包括参数的不确定性和输入的不确定性。在参数率定过程中,本文以单个参数率定为例,介绍了两种不确定性分析算法。实际应用中,参与率定的是几十个参数构成的参数组,观测值本身又是多个水质项目组成的向量,再加上模型的非线性特征极为突出,因此给参数的不确定性分析带来了相当的难度;由此引出,多个参数一起进行率定时应该采取什么样的策略,成为下一步的研究课题。

Eco-System-Based Lake Model and Its Uncertainty Analysis :A Case Study on Lake NanWan

Zeng Siyu, Du Pengfei, Ran Shenghong, J. Chen

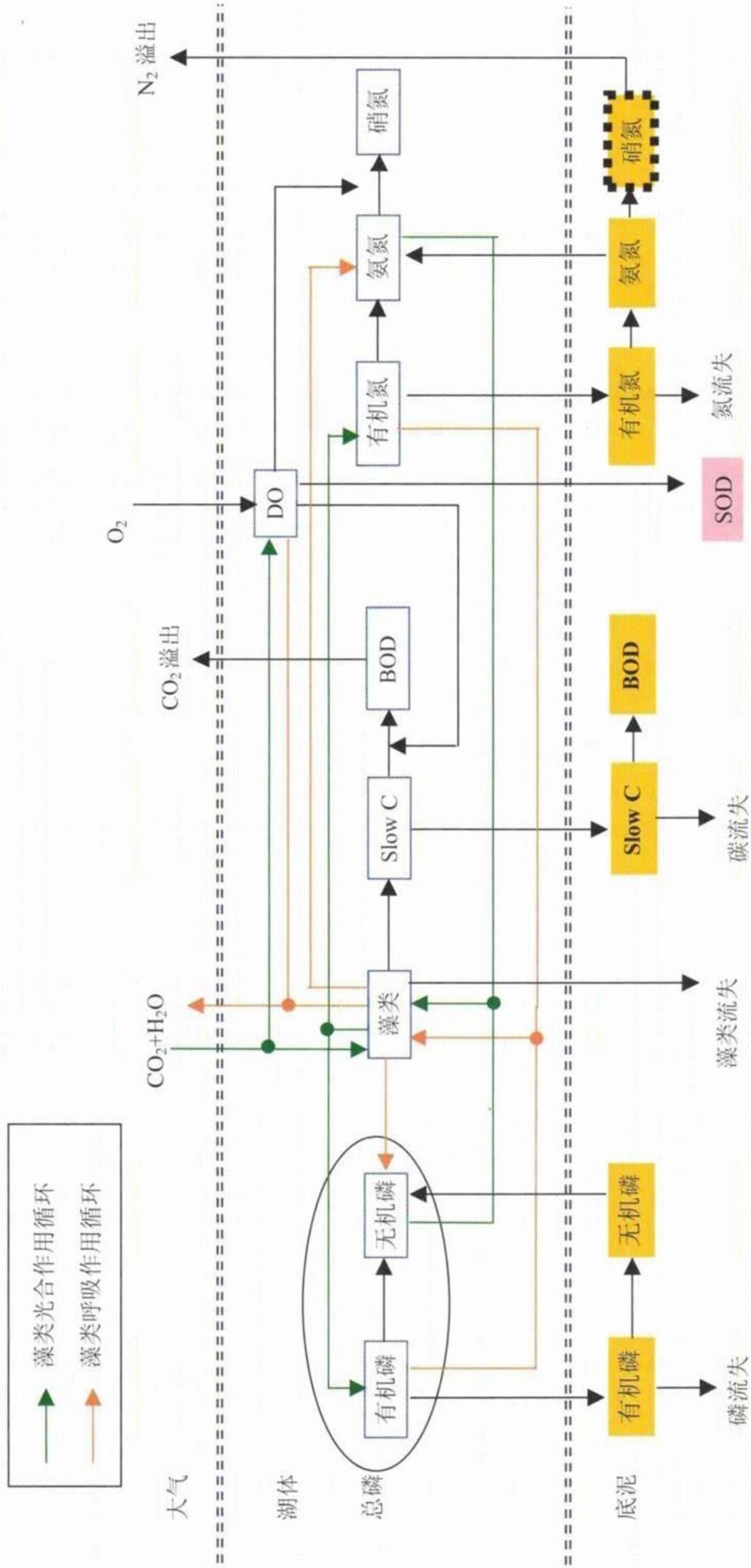
(Environmental Department, Tsinghua University, Beijing, 100084)

Wang Zhishi and Tang U Wa

(University of Macau, Faculty of Science and Technology, Macau, PRC)

Abstract: With the case study on Lake NanWan in Macao, the paper described the function and structure of a lake model based on eco-system. During the water quality simulation, two uncertainty analysis methods were attempted to calibrate model parameters, which was GLUE algorithm and HSY algorithm. According to uncertainty estimation of parameters, uncertainty transfer to simulation results was discussed. Furthermore, the uncertainty of pollution load as model input was shown.

Keywords: Lake Model; Uncertainty Analysis; GLUE Algorithm; HSY Algorithm



注：图中“硝氮”指亚硝酸盐氮与硝酸盐氮之和。

图 1 模型中各组分之间的主要关系

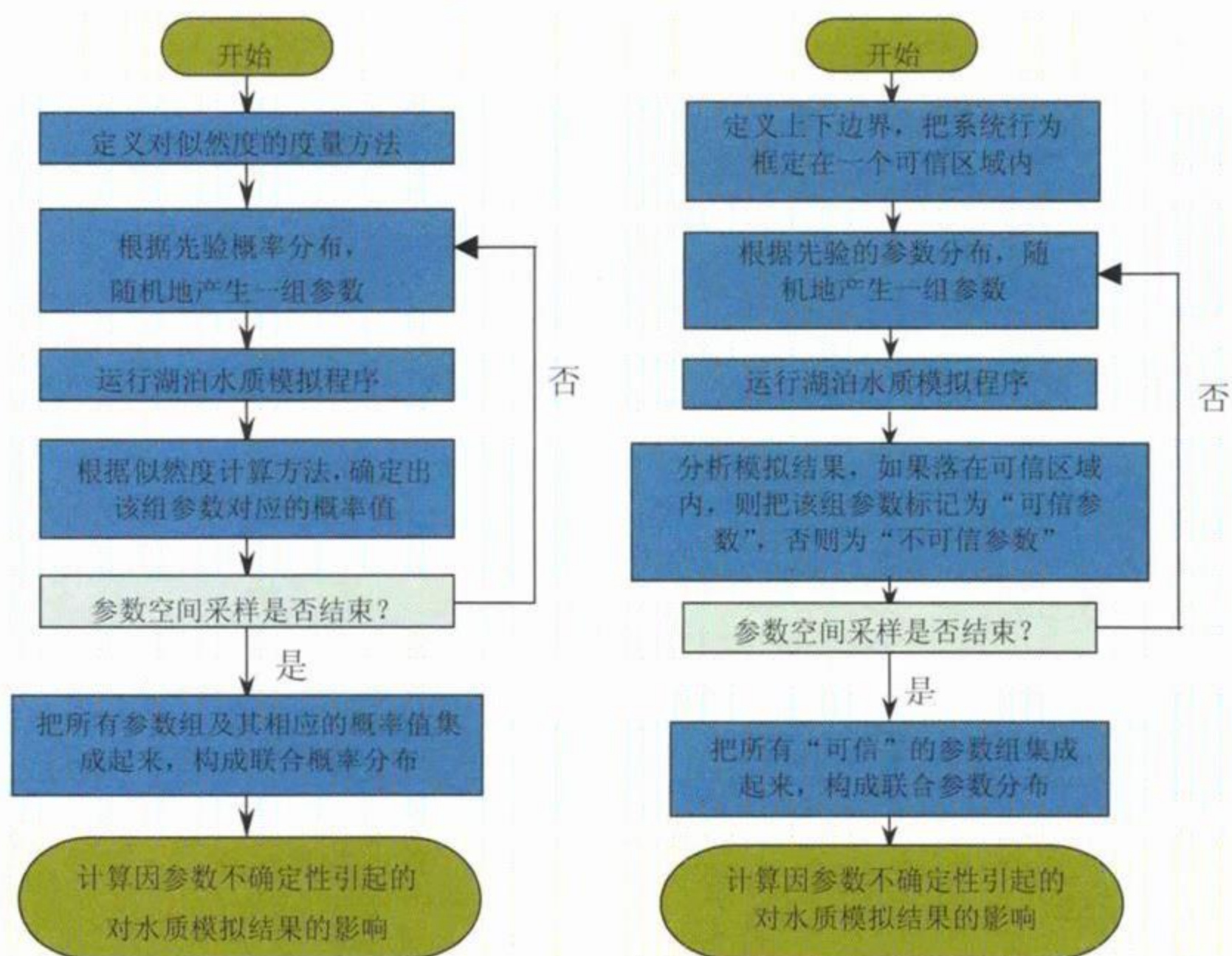


图 2 GLUE 算法的基本过程 (左) 和 HSY 算法基本过程 (右)

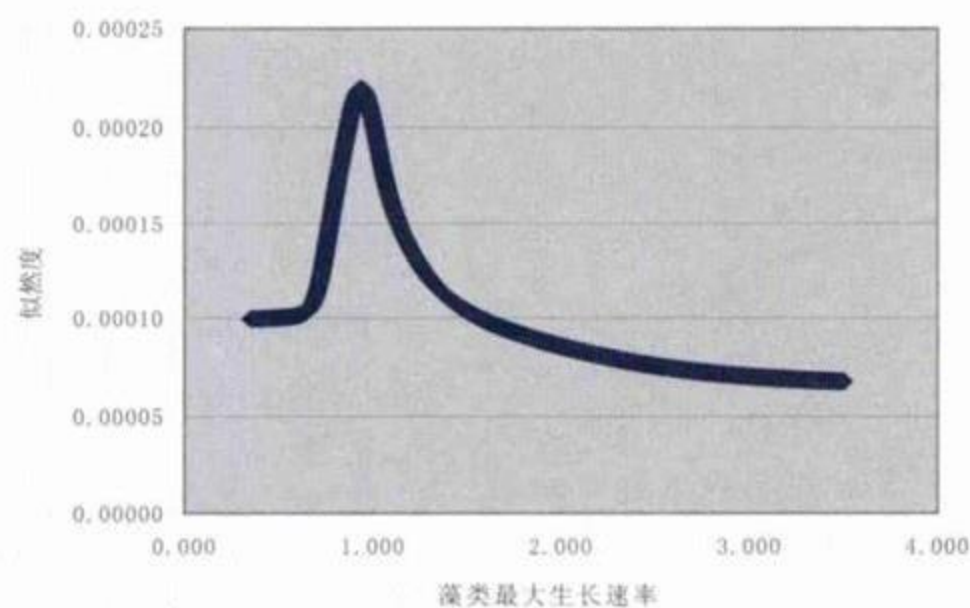


图 3 用 GLUE 算法率定参数 (藻类最大生长速率)

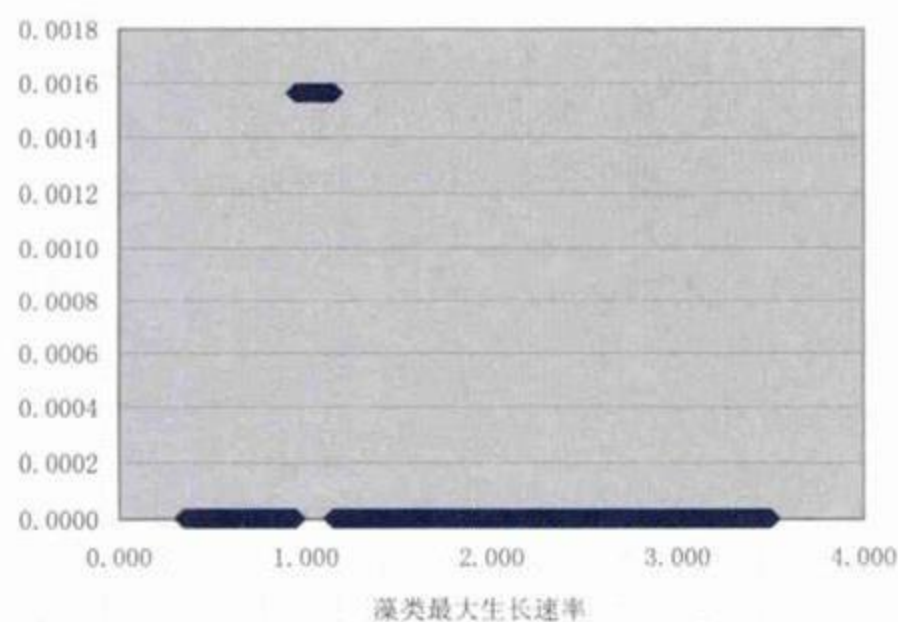


图 4 用 HSY 算法率定参数 (藻类最大生长速率)

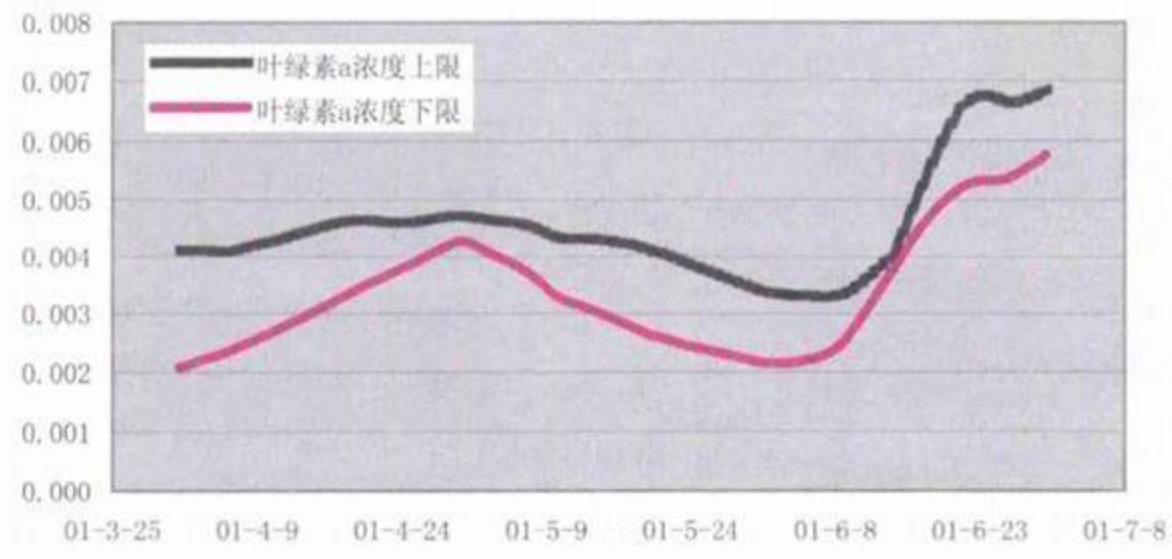


图 5 利用 HSY 算法获得的叶绿素 a 浓度不确定性分析结果

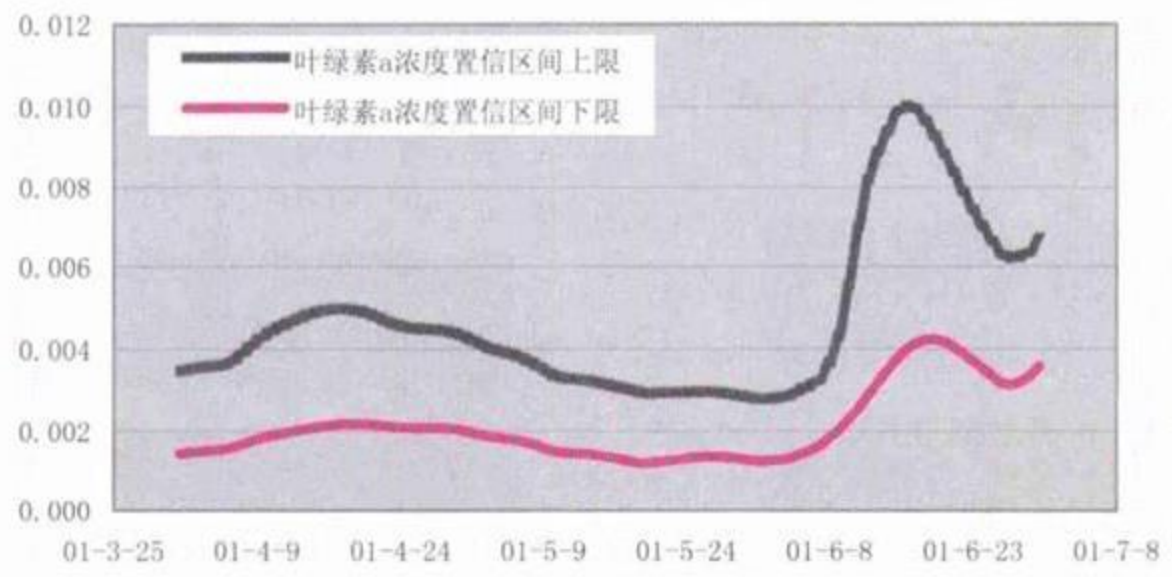


图 6 利用 GLUE 算法获得的叶绿素 a 浓度不确定性分析结果（80%置信度）

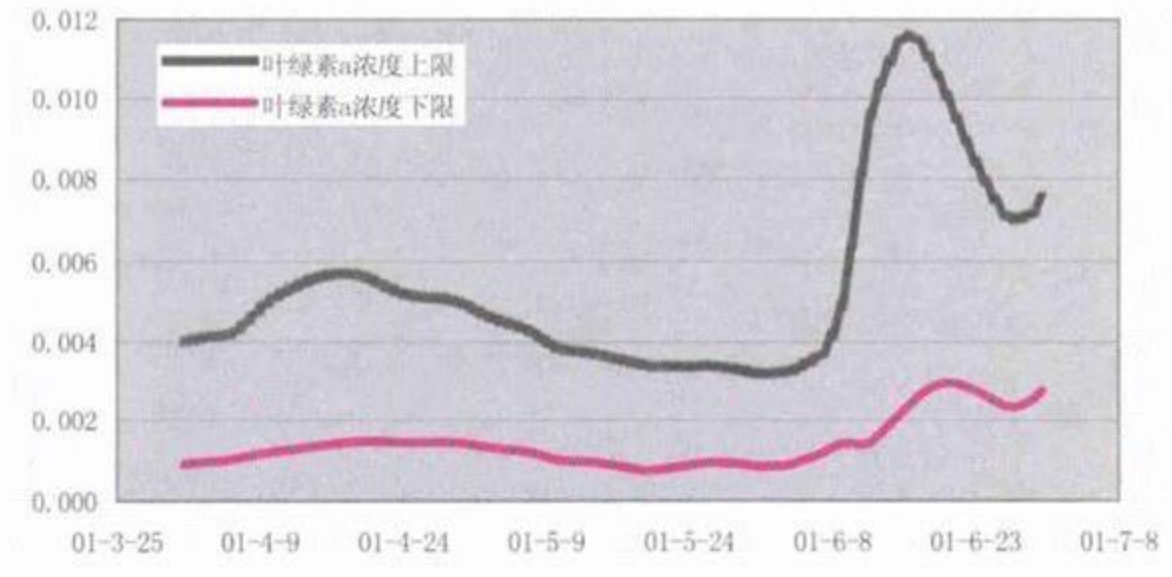


图 7 利用 GLUE 算法获得的叶绿素 a 浓度不确定性分析结果（90%置信度）

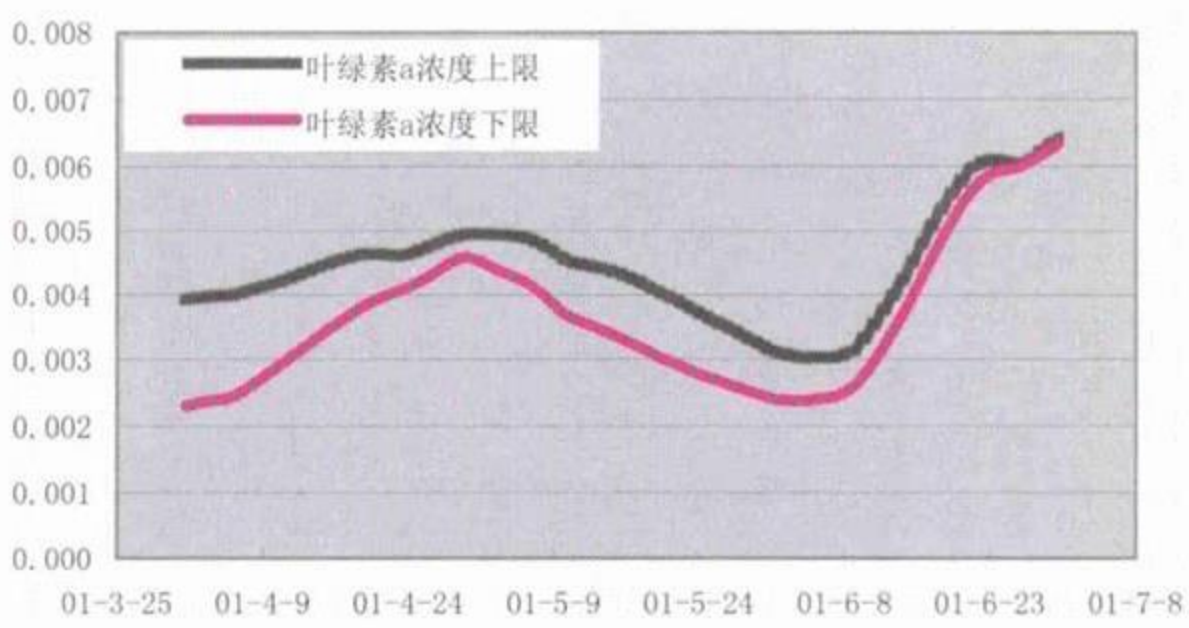


图 8 总磷负荷的不确定性对湖泊水质叶绿素 a 浓度的影响（HSY 算法）

评价管网水中异养菌二次生长潜力的首要指标——AOC

李 爽 张晓健

(清华大学环境科学与工程系, 北京 100084, 中国)

范晓军 刘慧敏 汪浩 叶慧娟

(澳门自来水有限公司)

摘要:本文研究了澳门管网水中异养菌二次生长和水质生物稳定性指标的相关关系,测定的主要项目包括:异养菌计数(HPC)、生物可同化有机碳(AOC)和生物可降解溶解性有机碳(BDOC)。结果表明:(1)澳门管网水基本属于生物稳定的饮用水。(2)澳门管网沿途 AOC 和 BDOC 的变化不明显,因此考察饮用水中生物稳定与否,可以以出厂水作为控制点进行研究。(3)降水量和水温对于 AOC 的季节间变化有很大影响;而原水水质、处理工艺和水温对 BDOC 的季节变化有很大影响。(4)由于管网水中 AOC 和 HPC 有明显相关关系,而 BDOC 与 HPC 间无明显相关关系,所以建议将 AOC 作为评价管网水中异养菌二次生长潜力的首要指标参数。

关键词:管网水, AOC, BDOC, 异养菌

1. 前言

合格的出厂水在管网中可能会发生细菌的再生长,造成饮用水的二次污染,影响水的嗅、味和色度,并可能引发致病菌的繁殖。早在 1930 年,美国 AWWA 供水委员会^[1]就报告了大肠杆菌二次生长的问题。从那时起的 70 年里,水工业一直就控制异养菌二次生长问题展开了不懈努力。消毒剂的投加,在一定程度上控制了管网中细菌的二次生长,但是消毒并不能完全解决该问题。多数研究表明即使保持管网中一定余氯,异养细菌在有机物存在下仍然会生长^{[2][3][4]}。

影响细菌在管网中再生长的因素很多,除了上文提到的消毒剂余量外,还包括营养基质、水力因素、温度、管材和管段腐蚀情况^{[5][6][7]}等。当一个管网系统相对固定时,从实际角度来看,人为能控制的因素主要是水中有机营养基质的浓度,为此提出了生物稳定性的概念,即饮用水中有机营养基质支持异养菌生长的潜力。饮用水生物稳定性高,则表明水中细菌生长所需的有机营养基质少,细菌不易在其中生长;反之则容易发生细菌再生长问题。

生物稳定性的评价指标主要有两种:生物可同化有机碳(AOC)和生物可降解溶解性有机碳(BDOC)。AOC 方法由荷兰 VanDerKooij 教授首创^[8],是在待测水样中接种特殊细菌,通过平板计数,测定其生长稳定期(stationary phase)细菌数,再比较该细菌在标准待测物(一般是乙酸)中生长稳定期的细菌数而求得水中可生化降解有机物的浓度。BDOC 是由比利时人 Pierre Servais 等人^[9]发明的方法,以接种原水中的混合菌种在待测水样中的培养,通过测定水样培养前后溶解性有机碳(DOC)的差值确定水中可生物降解有机物的值。这两类方法实质上都是生物检测(Bioassay)方法。AOC 是特定菌种转化为细菌生物量的那部分生物可降解有机物,测定的是异养菌的生长潜力;而 BDOC 则是水中能被多种土著异养菌降解的那部分有机物量,测定的是水处理中能被生物氧化的潜在的有机碳。一般认为 AOC 应包含在 BDOC 之中,但实际上二者之间没有一定的比例关系,互相独立,应用也各不相同。

目前美国等地应用 AOC 比较多,而法国等地应用 BDOC 比较多,但是尚未有明确的试验数据证明这两个指标哪个更适合于代表管网水中异养菌的二次生长情况。本文研究的目的就是:(1)评价澳门管网水的生物稳定性情况;(2)分析管网水中 AOC 和 BDOC 的变化;(3)研究 AOC、BDOC 与异养菌的相关关系,选出相关性较好的参数作为控制管网水中细菌二次生长

潜力的主要指标参数。

2. 试验条件和方法

2.1. 管网取样点的确定

2001 年夏季和秋季对澳门的 5 个管网点进行了水质生物稳定性的监测。由于澳门管网较短而且主要为环形供水, 所以沿主岛边缘选取了近可能长的三个管网点来代表管网中水质的变化, 分别为青州出厂水 (99 号, 编号源自澳门自来水公司取水点, 下同), 管网中部点 (109 号) 和管网末梢点 (39 号)。另外还选取了两个水质较差 (主要根据细菌指标) 的管网点进行测定, 即路环水警分所 (61 号) 和同善堂中学附属幼稚园 (111 号)。取水点如图 1 所示。

2.2. 试验仪器的准备

所有玻璃仪器都要保证无碳化。酸洗后浸泡过夜, 然后分别用自来水、蒸馏水和高纯水润洗三次。通过 550℃ 下加热 2 小时来去除痕量碳。采样瓶加入硫代硫酸钠中止余氯。

2.3. AOC 测定方法

40mL 水样经 70℃ 30 分钟水浴进行巴氏消毒, 冷却至室温, 同时接种 P17 和 NOX, 2—25℃ 下培养 2—3 天, 再对培养液进行平板计数, 计算水样中的 AOC 浓度。产率系数采用在澳门自来水公司测得数据: $Y(P17) = 4.3 \times 10^6 \text{cfu}/\mu\text{g}$, $Y(\text{NOX}) = 0.7 \times 10^6 \text{cfu}/\mu\text{g}$ 。

2.4. BDOC 测定方法

测定方法采用的是 1986 年 Joret 和 Levi 提出方法^[10]的修改版, 接种细菌是来自石英砂滤池的附着细菌。取 100g 活化后的石英砂放入锥形瓶中, 加入 300mL 已中止余氯的待测水样, 测定加砂前后的 DOC 差值 (超过 0.2mg/L 的作废), 并通过空白, 空白加砂和标准加砂水样来进行质量控制, 标准为 2mg/L 的乙酸钠水样。每天同一时间测定水样的 DOC 值, 一般在 5 到 10 天内可降至最低, 计算前后的 DOC 差值即为 BDOC 的数值。试验在室温下进行, 注意避光。

2.5. TOC 测定

采用岛津 5000A TOC 仪进行测定。

2.6. 异养菌 (HPC) 测定方法

使用铺平板技术, 22—25℃ 下在 R2A 琼脂上培养 7 天计数。平行样 3 个。

3. 试验结果和分析

3.1. 澳门管网水中 AOC 的变化

图 2 所示为 AOC 在澳门管网水中 6—10 月份的变化情况。一般认为加氯情况下保持生物稳定的管网水 AOC 范围为 50 到 100 $\mu\text{g}/\text{L}$, 而澳门管网水中的 AOC 在夏季气温最高的时候也不超过 70 $\mu\text{g}/\text{L}$, 其他月份基本在 30 到 50 $\mu\text{g}/\text{L}$ 之间, 因此可以认为, 澳门饮用水基本属于生物稳定的饮用水。

从出厂水到管网末梢之间, AOC 变化幅度不大, 最大不超过 14 $\mu\text{g}/\text{L}$ 。这主要是因为澳门管网比较短, 所以管网中的 AOC 变化不明显, 因此考察饮用水中生物稳定与否, 可以以出厂水作为控制点进行研究。

比较 AOC 季节间的变化规律, 可通过图 2 的月平均值 (见图中虚线) 来看。六七八月份

的平均值非常接近，九月份升高至 56μg/L，十月份则降至 27μg/L。管网水中 AOC 受到很多因素的影响，如原水水质、水温、降雨量、余氯量等多种因素的影响，下面就这些参数列表进行分析，见表 1。

表 1 澳门 6—10 月份水温、降雨量和出厂水余氯数据

月份	水温(℃)	降雨量(mm)	出厂水余氯(mg/L)
6	28.0	729.8	0.9
7	28.0	666.9	0.8
8	30.5	191.6	0.7
9	27.3	/	1.0
10	26.0	/	0.9

从表中可以看到，降雨量对于原水中 AOC 的含量有显著影响，由于 6、7 月份是澳门降水高峰期，导致原水中有机物浓度下降，后续月份(即 7、8 月份)水源水中 AOC 值逐渐降低，8 月份已降至 28.6μg/L。不过由于 7、8 月份水温比较高，AOC 含量会相应上升，综合以上两种因素，这两个月份出厂水的 AOC 值仍然和 6 月份的非常接近。由于 8 月份降水较少，导致温度仍较高的 9 月份水源水 AOC 值偏高，继而出厂水中 AOC 值也很高，可以看出澳门自来水公司因此投加的消毒剂量增加，导致出厂水余氯达到最高 1.0mg/L。10 月份温度下降，水中 AOC 含量开始明显低于前几个月。

3.2. BDOC 在给水管网中的变化

图 3 所示为 BDOC 在澳门给水管网中 6—10 月份的变化情况。从出厂水到管网末梢之间，BDOC 变化幅度不大，一般均在 0.2mg/L 以下。这主要是因为澳门管网比较短，所以管网中的 BDOC 变化不明显。

比较季节间的变化规律，可通过图 3 的月平均值来看。五六月月份持续上升至 0.47mg/L，八月份数据和七月份数据接近，九月份下降至 0.39mg/L，十月份上升至 0.60g/L。

分析管网水中 BDOC 的季节变化，可以看出原水水质、处理工艺和水温对其变化有很大影响。五六七月持续上升是由于气温上升所致；八月与七月持平原因是原水水质明显好于七月，抵消了温度影响；九月温度下降 BDOC 也随之下降，至于十月反常的增高可能是因为原水中 BDOC 较高，而且水厂工艺对它基本无去除所致。

3.3. 管网水中 HPC 与 TOC、BDOC 和 AOC 的相关性

图 4 所示为 HPC 和 TOC 的相关关系图，可以看出二者无任何相关关系。HPC 和 BDOC 也无明显的相关关系，如图 5 所示。可见 TOC 和 BDOC 都不适合用来指示管网水中异养菌的生长潜力。而澳门管网水中 HPC 和 AOC 却有明显的相关关系，如图 6 所示，其结果和 van der Kooij 的研究结果^[11]类似，如图 7 所示。

3.4. 评价管网水异养菌二次生长潜力的首要指标

AOC 和 BDOC 同为生物稳定性的指标，二者的定义不同，测定方法原理不同。究竟哪一个指标更适合用于评价管网水中异养菌二次生长的潜力，还没有定论。

笔者对澳门水源水进行膜过滤分析，发现二者所代表的有机物分子量也有明显差异^[12]，如图 8 和图 9 所示。由图 8 可以看出，AOC 随着分子量的增加没有明显的梯级效果，三种水源水在小于 1000，3000，10000，100000 表观分子量之下的 AOC 含量变化均在方法精密度范围允许之内，彼此之间没有显著性差异。因此推测 AOC 主要与分子量小于 1000 的有机物相关，此结论与其他研究者^[13]结论一致。由图 9 可以看出，分子量 10000 的超滤膜对 BDOC 的去除率低于 45%，分子量 1000 的超滤膜对其去除率则在 49-80%，这意味着 BDOC 不是仅仅主要与分子量小于 1000 的有机物相关，与 AOC 不同，在 1000—10000 之间存在着相当部分有机物与 BDOC 相关。

管网水中异养菌计数结果和 AOC 的相关关系较好，原因可能是因为其中异养菌利用的主要是低于 1000 道尔顿的小分子量有机物，对于高于 1000 道尔顿的有机物利用效率较低。另外，AOC 是采用特定细菌 (P17 和 NOX) 进行测定的，这两种细菌广泛存在于管网水中；而 BDOC 采用的是水源水中的土著混合菌种，由于处理工艺的去除，和管网水中的异养菌种类差别较大，也可能导致与 HPC 相关关系的差异。

因此，根据澳门饮用水的情况，与 BDOC 比较而言，AOC 更适于作为评价管网水中异养菌二次生长潜力的指示参数。

4. 结论

- (1) 澳门管网水中的 AOC 在夏季气温最高的时候也不超过 70μg/L，其他月份基本在 30-50μg/L 之间，基本属于生物稳定的饮用水。
- (2) 澳门管网水沿途 AOC 变化不明显，因此考察饮用水中生物稳定与否，可以以出厂水作为控制点进行研究。
- (3) 降水量和水温对于 AOC 的季节间变化有很大影响，由于澳门 6、7 月份为降水高峰期，所以 AOC 的夏季最高点向后推移至 9 月份，比前几月份增加了 40% 左右。
- (4) 从出厂水到管网末梢之间，BDOC 变化幅度不大，一般均在 0.2mg/L 以下。原水水质、处理工艺和水温对其季节变化有很大影响。
- (5) 由于管网水中 HPC 和 AOC 间有明显相关关系，而 HPC 与 BDOC 间则无明显相关关系，所以评价管网水中异养菌二次生长潜力的指示参数，AOC 比 BDOC 更适合。



图 1 澳门管网取样点示意图

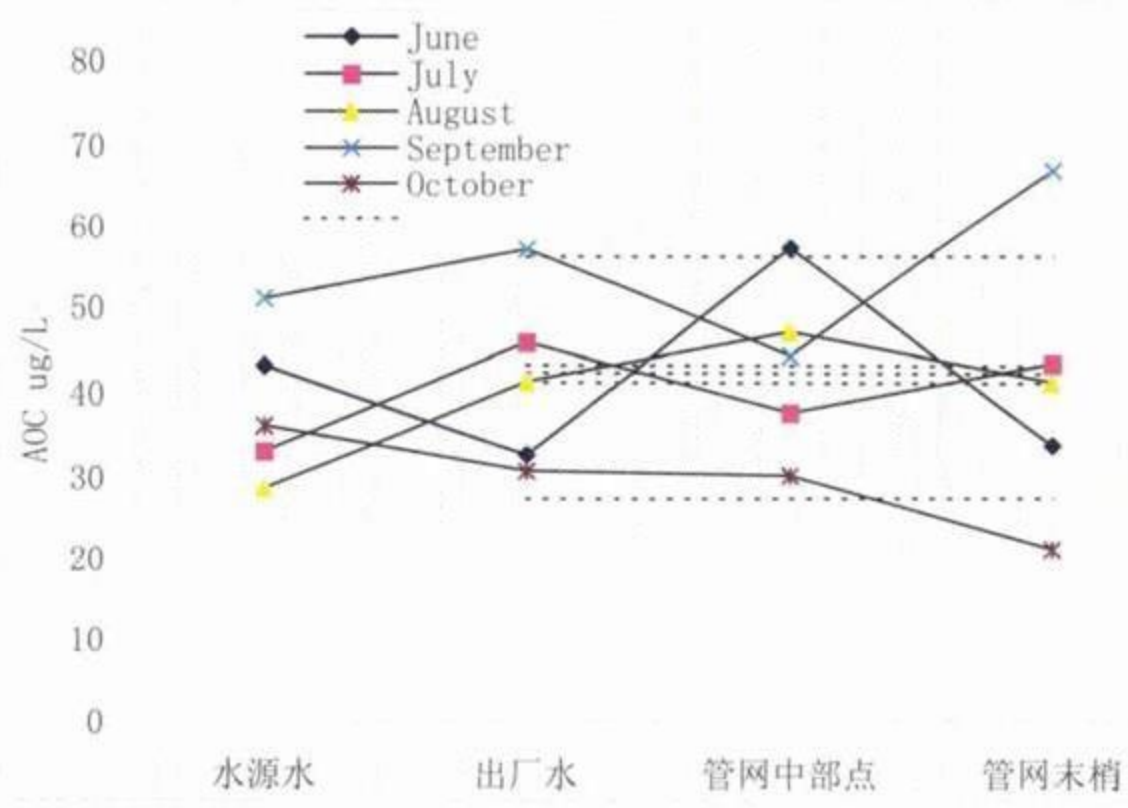


图 2 AOC 在澳门管网水中的变化情况

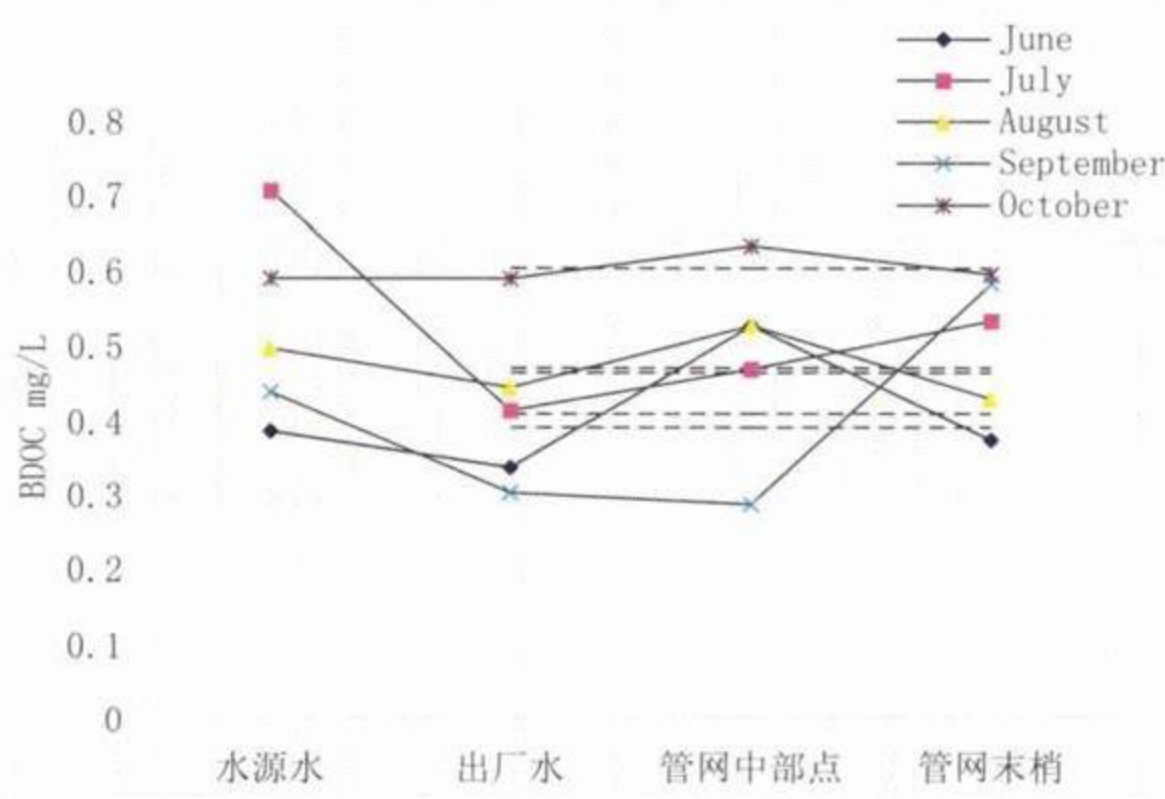


图 3 BDOC 在澳门管网水中的变化情况

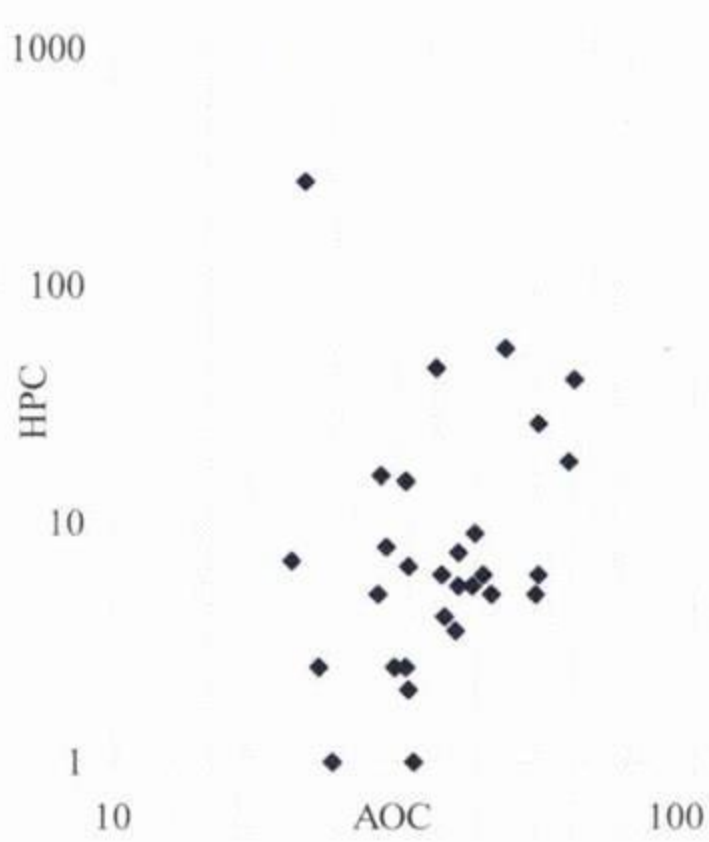


图 4 澳门管网水中 HPC 和 AOC 的相关关系图

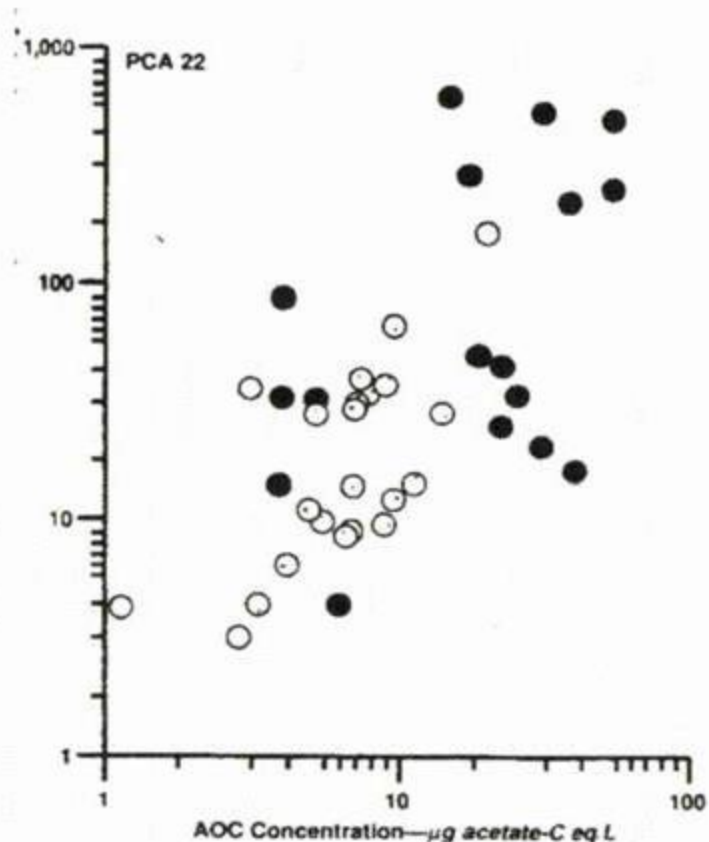


图 5 van der Kooij 研究中 HPC 和 AOC 的相关关系图

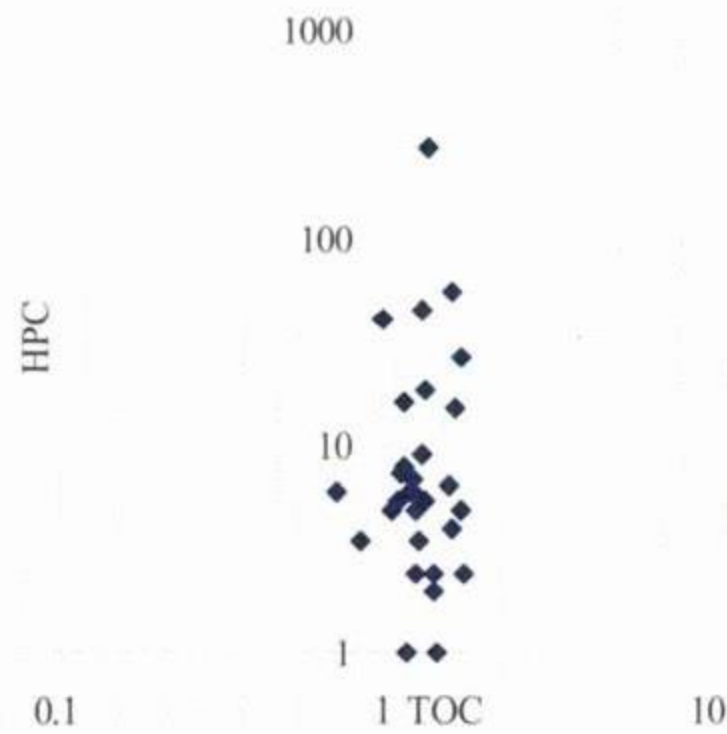


图 6. 澳门管网水中 HPC 和 TOC 的相关关系图

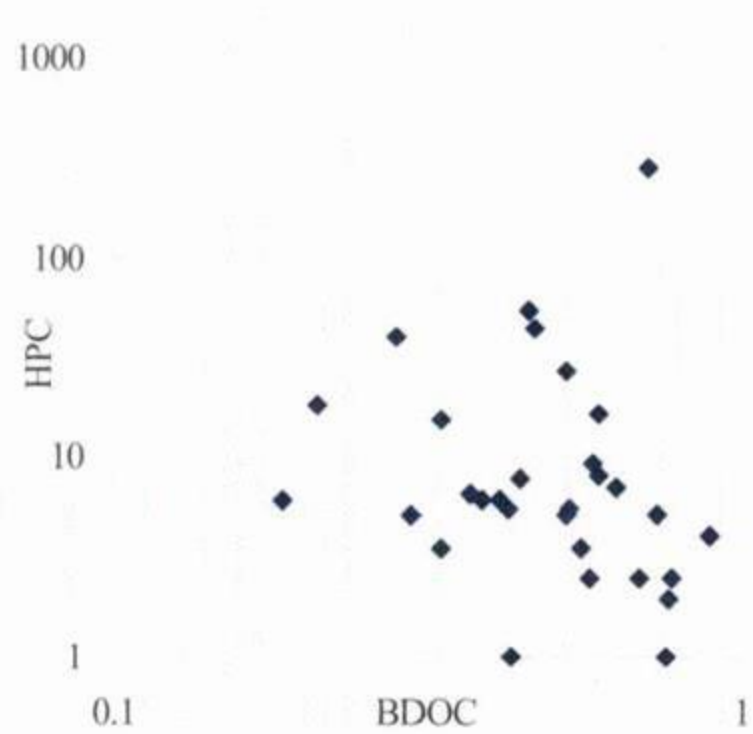


图 7 澳门管网水中 HPC 和 BDOC 的相关关系图

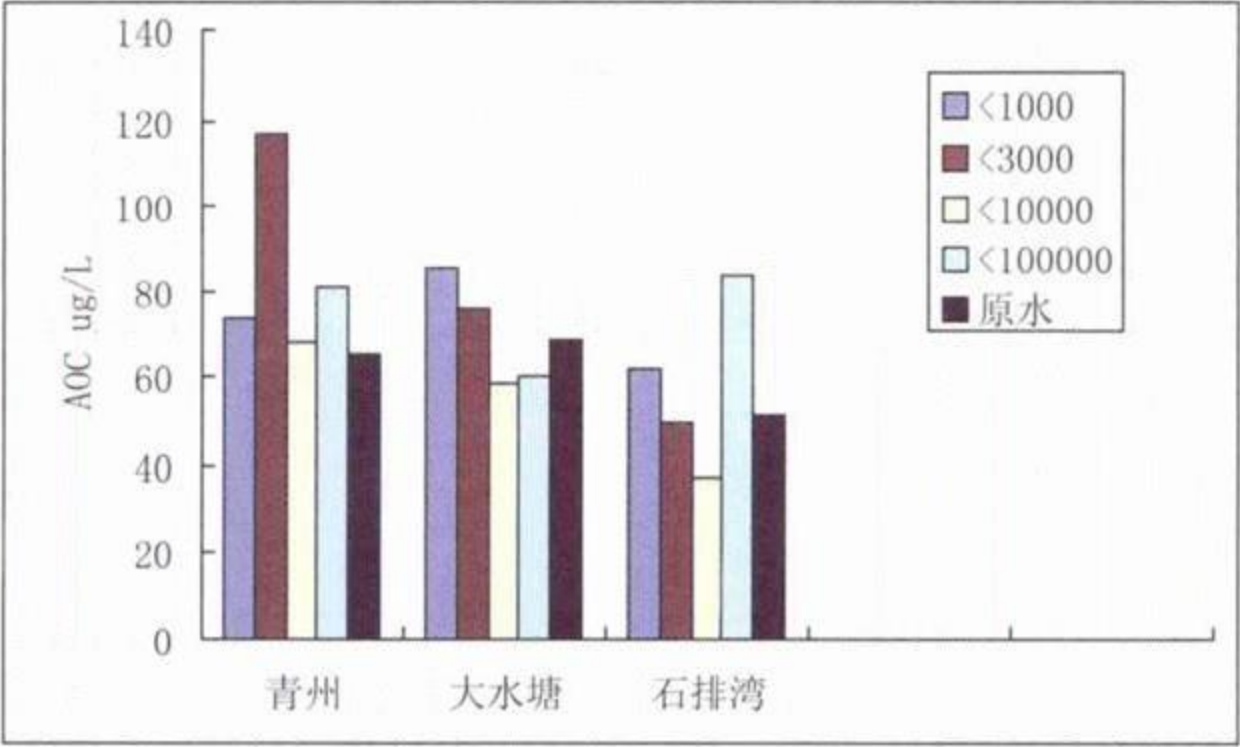


图 8 澳门水源水中小于某一表观分子量有机物的 AOC

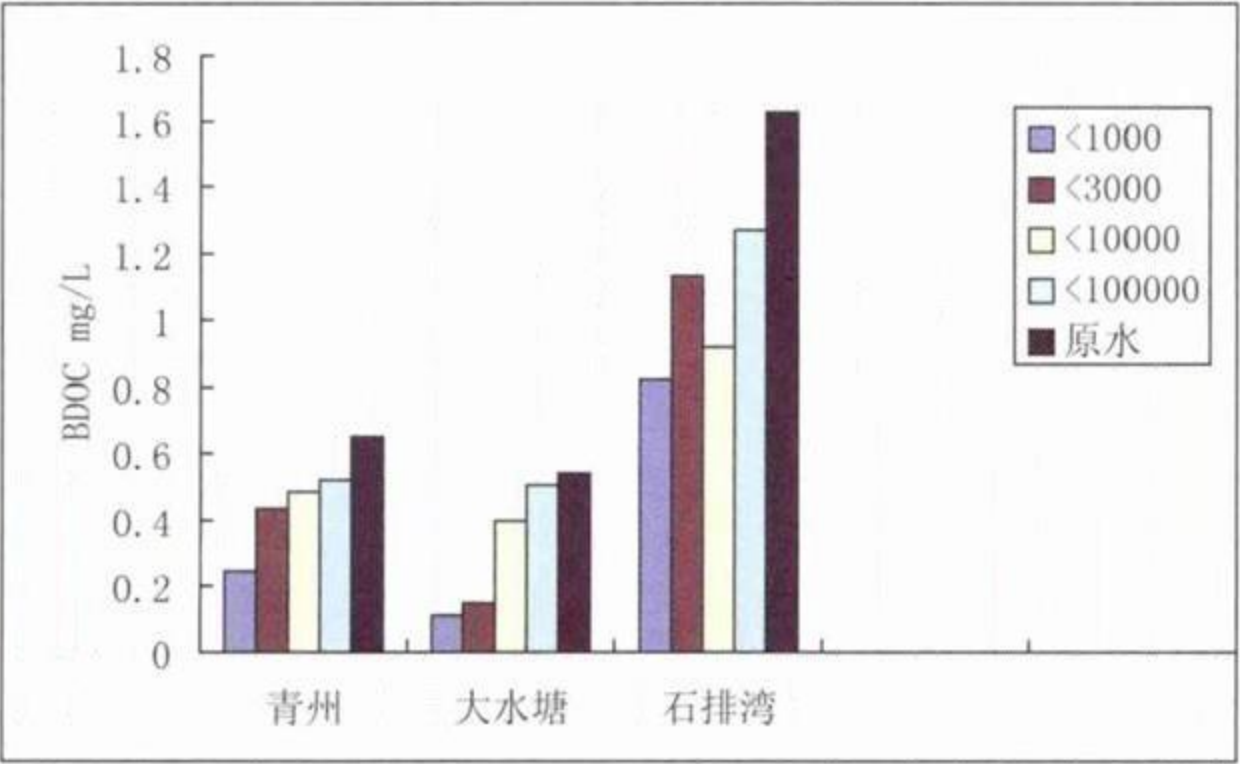


图 9 澳门水源水中小于某一表观分子量有机物的 BDOC

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Assimilable Organic Carbon as an Indicator of Bacteria Regrowth Potential in Drinking Water

Li Shuang and Zhang Xiaojian

Department of Environmental Science and Engineering in Tsinghua University, Beijing 100084

Fan Xiaojun, Lao Wai Mom, Wong Hou and Ye Huijuan

The Macao Water Supply Company Ltd.

Abstract: Assimilable organic carbon(AOC) and biodegradable dissolved organic carbon(BDOC) were measured in Macao drinking waters. The results show: (1) Macao drinking water has, or approximates to, biological stability according to the AOC analysis. (2) AOC and BDOC fluctuations appear small in Macao distribution system. (3) AOC seasonal changes are influenced by precipitation quantity and water temperature, while BDOC seasonal changes are influenced by raw water quality, treatment process and water temperature. (4) AOC has obvious correlation with heterotrophic bacteria colonies(HPC) in distribution waters, while BDOC has no correlation with HPC, therefore, it can be proposed that AOC be used as an preferential indicator of bacteria regrowth potential in drinking water.

Keywords: drinking water; assimilable organic carbon; biodegradable dissolved organic carbon; heterotrophic bacteria colonies.

澳门半岛城市噪声模拟与控制研究

白松涛, 傅立新

(清华大学环境工程和科学系)

王志石, 谭伟文, 陈海英, 邓宇华

(澳门市政厅和澳门大学)

摘要: 交通噪声是澳门半岛最主要的环境噪声源。本文在国际上通用的交通噪声预测模型 STAMSON 的基础上, 根据澳门半岛封闭型道路的特征, 建立了封闭型道路交通噪声计算机预测模式, 经过实测数据的验证, 模型预测结果达到了令人满意的精度要求。利用建立的交通噪声预报模式, 计算了澳门半岛不同类型道路的交通噪声水平, 定量评价了道路交通噪声的污染状况。最后, 根据不同功能区监测的噪声水平, 分析主要噪声源对不同区域的环境噪声影响以及澳门半岛交通密度高的特点, 提出了新的环境噪声标准建议, 并给出了逐步改善环境噪声的主要措施。

关键词: STAMSON、交通噪声、噪声模拟, 功能区

1. 前言

由于澳门半岛是一个东南至西北窄, 东北至西南宽的狭长半岛, 岛上的道路相当狭窄且无序, 许多路段坡陡弯急。另一方面, 随着社会经济的发展, 澳门市的车流量明显增大, 导致道路两旁的噪声显著增加, 严重影响到了澳门市民的起居生活。这次对澳门半岛的调研的内容包含了以下四个方面: 城市噪声监测评价、交通噪声模拟分析、GIS 支持平台和噪声标准与控制对策。本文中将对城市噪声监测和交通噪声模拟进行重点的分析与研究, 同时结合澳门城市噪声现状提出新的噪声标准。

2. 噪声预测模式

在城市各种噪声源中, 无论从污染面或污染强度看, 交通噪声都是最重要的噪声源, 因此对于城市交通噪声的预测成为国内外噪声研究的一个重点。在国外提出了多种道路交通噪声预测模型, 比如美国的 FHWA 高速公路交通噪声预测模型、英国的 CRTN88、多层递阶模型等等。而在国内, 由于我国城市交通情况极为复杂, 道路狭窄, 而且车辆类型多, 混合行驶, 车速较慢, 旧车比例高, 所以国内一些研究人员对上述模型进行了相应的改进或者是建立符合当地情况的预测模型。

在本次研究中采用了加拿大的 STAMSON 模型, 该模型采用了以美国 FHWA 公路噪声预测模式为基础的 ORNAMENT (Ontario Road Noise Analysis Method for Environment and Transportation) 和 STEAM (Sound from Trains Environment Analysis Method) 两种方法, 经参数修正后可以模拟预测澳门的交通噪声水平。

FHWA 公路噪声预测模式适用范围大, 而且精度高 (在噪声接受器在 15m 的距离下误差在 1.64dB 内)。FHWA 模式将道路上汽车按车流进行分类, 先求出某一类车辆的小时等效声级, 公式如下:

$$Leq(h_i) = \overline{L_{oe,i}} + 10 \log \left(\frac{N_i \pi D_0}{S_i T} \right) + 10 \log \left(\frac{D_0}{D} \right)^{1+\alpha} + 10 \log \left[\frac{\psi_\alpha(\varphi_1, \varphi_2)}{\pi} \right] + \Delta S - 30 + B \quad (1)$$

式中: $Leq(h_i)$ ——第 i 类车辆 h 小时的 Leq , $dB(A)$;

- $L_{oe, i}$ ——第 i 类车辆的参考能量平均辐射声级, $dB(A)$;
- N_i ——在制定时间 T 内通过某预测点的第 i 类车流量, 辆;
- D_0 ——测量车辆辐射声级的参考位置距离, m ;
- D ——从车道中心到预测点的垂直距离, m ;
- S_i ——第 i 类车辆的平均车速, km/h ;
- T ——计算等效声级的时间, h ;
- α ——地面覆盖系数, 取决于现场地面条件;
- ψ_α ——代表有限长路段修正的函数符号, 其中 φ_1 、 φ_2 为预测点到有限长路段两端的张角, rad ;
- ΔS ——由遮挡物引起的衰减量, $dB(A)$;
- B ——道路封闭程度修正因子, $dB(A)$ 。

针对澳门半岛主要为封闭型街道的特点, 本文在上述公式的基础上, 增加了一项街道封闭程度修正因子 B 。该因子根据不同街道的高宽比(两侧建筑物高度与街道宽度之比)来确定由于建筑反射而引起的街道噪声增加量。计算公式如下:

$$B = \log \left[\frac{2}{\pi} \arctg \frac{b}{2h} \right]$$

(2)

式中: b 和 h 分别表示街道的宽度和周围房屋的高度。

混合车流模式的等效声级是将各类车流等效声级叠加到一起, 如果将车流分成轿车、中型卡车和重型卡车三类, 那么总车流等效声级为

$$Leq(T) = 10 \log(10^{0.1Leq(h1)} + 10^{0.1Leq(h2)} + 10^{0.1Leq(h3)})$$

(3)

其中 $Leq(h_i)$ ($i=1,2,3$) 分别对应轿车、中型卡车和重型卡车的小时等效声级。

3. 数据调查

数据清单中包含了交通流量、车辆行驶速度、道路坡度以及根据澳门半岛地区地理或道路特征的一些设置参数。

澳门大学负责调查了澳门半岛主要 44 条马路的车流量和行驶速度, 并根据实际情况将车辆分为了摩托车、私家车、小型货车、出租车、卡车和公共汽车六种车型。不同类型的车辆形成的噪声级具体如表 1。该表给出了各种车型的噪声级与轿车噪声级的比值, 以及相应的分类。但是由于在 STAMSON 模型中, 只是将汽车分为了轿车、中型卡车和重型卡车。根据表 1, 我们将私家车、小型货车和出租车归为轿车, 摩托车和公共汽车归为中型卡车, 卡车归为重型卡车。

表 1 在平均速度下不同类型车辆形成的噪声级 (单位: dB)

	AU	LT	MT	HT	MC	BU	MB
噪声水平	65.00	67.50	72.50	74.60	71.00	71.00	71.00
与轿车的比值	1.00	1.04	1.12	1.14	1.09	1.09	1.09
车型分类	AU	LT	MT	HT	(MC+BU+MB)		

表中: AU——轿车 (automobile);
LT——轻型卡车 (light truck);
MT——中型卡车 (medium truck);

- HT——重型卡车 (heavy truck);
- MC——摩托车 (motorcycles);
- BU——公共汽车 (bus);
- MB——小公共汽车 (minibus)。

根据澳门地区车辆行使的特点，将速度分为了三个档次：0 至 20km/hr、20 至 40km/hr 和 40km/hr 以上。由于澳门交通拥挤，所以除了少数的几条干道（友谊大马路和慕士拉大马路）外，其他街道的车辆行驶速度都在 40km/hr 以下。所以在使用 STAMSON 模型进行计算的时候，单独对这两条街道进行了速度取值（其中友谊大马路速度取值为 55km/hr，慕士拉大马路速度取值为 50km/hr）模拟，其余街道都是采用 40km/hr 作为最大速度来模拟计算。另外由于澳门地区多山路，所以道路坡度也是进行交通噪声模拟的一个重要指标。将坡度分为了 0 至 5 度、5 至 15 度和 15 至 30 度三个档次，然后依次对每条街道进行调查。除了上面提到了车流量、道路坡度和行驶速度的三个参数以外，还有一些其他的参数需要根据澳门地区的地理或者道路特征进行设定。

4. 交通噪声现状模拟

利用在数据清单中建立的数值，代入到 STAMSON 模型就可以得到澳门半岛 44 条主要街道的模拟交通噪声级。在模拟出街道的等效噪声级后，就可以利用这些数值来计算监测点的等效噪声级。计算的过程中主要采用了以下几个方法：对于位于一条街道旁边的监测点直接采用模拟的街道等效噪声级；位于交叉路口的监测点利用其中两条主要街道的交通噪声进行能量叠加，进行模拟计算；对于行人功能区的监测点不能采用街道的交通噪声进行模拟，比如监测点 4（白鸽巢）和监测点 6（议事亭前地），这些地方本身的特征与街道有很大的差异，模拟值与实际监测值会有较大的误差。具体计算结果如表 2 和表 3 所示。

表 2 白天各时段实际监测 Leq 值和模拟 Leq 值（单位：dB）

监测点	模拟值				实际监测值			
	D1	D2	D3	D4	D1	D2	D3	D4
监测点 1	73.4	74.4	72.7	73.9	73.6	74.3	73.9	73.9
监测点 3	70.2	71.3	69.6	70.7	71.3	71.3	70.4	72.1
监测点 5*	74.8	73.6	74.5	74.9	74.8	74.8	74.3	75.1
监测点 7	74.4	74.4	73.7	74.8	75.0	75.1	75.3	75.6
监测点 8*	74.6	76.2	74.0	75.1	75.1	75.7	75.5	75.6
监测点 11*	75.7	77.3	75.3	76.2	76.2	76.5	76.2	76.4
监测点 12*	70.3	71.8	69.9	70.8	70.2	70.1	70.4	70.8
监测点 18*	70.6	71.1	71.8	69.7	70.2	69.4	69.6	70.4

注：带*号的监测点表示该监测点是交叉路口

表 3 夜间各时段和平均的实际监测 Leq 值和模拟 Leq 值（单位：dB）

监测点	模拟值			实际监测值			平均 差值
	N1	N4	均值	N1	N4	均值	
监测点 1	71.1	66.4	73.6	69.8	63.2	70.2	0.3
监测点 3	67.5	63.2	70.5	68.4	62.6	68.4	0.8
监测点 5*	73.4	68.6	74.5	73.4	58.6	70.4	0.3
监测点 7	72.0	67.6	74.3	73.5	68.0	73.1	1.0
监测点 8*	71.3	68.5	75.0	73.4	68.8	73.3	0.5
监测点 11*	73.5	68.6	76.1	75.0	67.8	73.9	0.2
监测点 12*	66.7	64.4	70.7	69.0	63.2	68.3	-0.3
监测点 18*	68.0	59.7	70.8	66.5	62.5	67.2	-0.9

注：带*号的监测点表示该监测点是交叉路口

将经过街道封闭程度修正后的模拟值与监测值比较，可以看出两者的符合程度很好，均值误差最大值只有 1dB，在各时段模拟值与实际监测值的比较中最大误差也不会超过 1.5dB。

图 1 是利用 STAMSON 模型模拟澳门半岛 44 条主要街道的等效噪声级 GIS 表现图。按照中国国家环境噪声标准 GB3096-95 中的规定“交通干线两侧昼间噪声不能高于 70dB”，在图 1 中按噪声级水平将 44 条街道分为三个档次，超标的街道有 9 条，占有所有街道的 20%；另外一方面相对噪声级较低的街道也有 9 条。此外，还发现噪声较高的交通区也是重型卡车流量相对较大的地区，反映出了重型卡车流量对一个地区噪声影响的重要性。



图 1 澳门半岛主要街道模拟噪声级 GIS 表现图

5. 澳门城市噪声评价

澳门地区按照行使的功能不同，可以分为交通、住宅、商业、休憩和工业五种不同的功能区。但是由于澳门地区错综重叠，因而每一个地区都不是行使单一的社会功能，而是集中了两种或者三种社会功能，甚至少数地区还具有了四种功能。综合澳门半岛上各个地区的特点，可以将半岛分为以下八种复合社区：休憩区、工业住宅区、交通住宅区、交通区、混合区、商住交通区、商业交通区和工业交通区。图 2 就是澳门半岛功能区的地理分布图，其中图例右边的数字表示在网格分布图中对应该种复合功能区的数量。从图 2 的地理分布图上，反映出澳门半岛主要是以交通住宅区（占据 14 个网格）和交通区（占据 16 个网格）为主，其他的功能区相对较少。

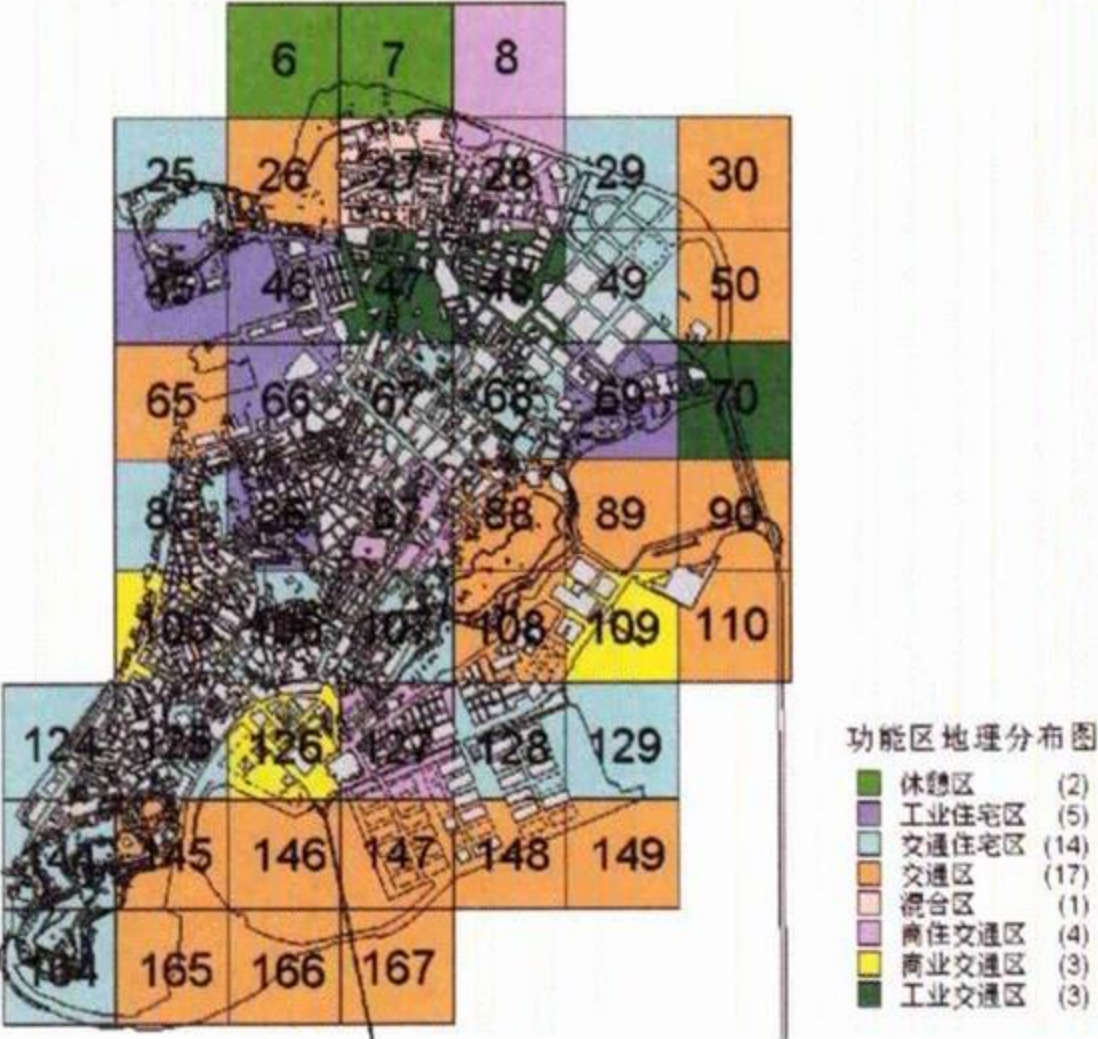


图 2. 澳门半岛地区复合功能区的地理分布图

根据对于49个网格2000年监测的数据,下表是关于八个功能区噪声特征参数的计算结果。

表 4 八个复合功能区的噪声特征参数

功能区	昼间			夜间		
	L_{eq}	$L_{max}-L_{eq}$	$L_{eq}-L_{95}$	L_{eq}	$L_{max}-L_{eq}$	$L_{eq}-L_{95}$
休憩区	60.37	18.37	6.43	52.23	16.27	9.37
工业住宅区	65.48	20.54	7.92	53.06	20.52	6.50
交通住宅区	66.17	19.41	9.26	55.97	18.09	8.70
交通区	65.48	17.21	8.91	57.07	18.64	9.22
混合区	70.40	13.20	6.40	57.70	12.50	5.10
商住交通区	68.65	16.40	6.55	60.68	16.95	6.18
商业交通区	70.43	15.07	6.30	61.53	15.80	5.40
工业交通区	74.20	16.05	6.80	64.30	17.15	10.80

根据噪声水平将八个功能区分两类,其中前四个复合功能区(休憩区、工业住宅区、交通住宅区和交通区),这些功能区的特点是交通流量少,或者交通量较大,但商业密度低,所以这些地区噪声水平相对较低,昼夜的噪声水平都在国标的规定范围以内;而后复合四个功能区(混合区、商住交通区、商业交通区和工业交通区)商业活动频繁,车流量大,造成了这些地区的噪声水平高,甚至有些地区的平均噪声水平高出国标 10dB 以上。(在这里需要指出的是工业交通区噪声水平高的原因并不是由于商业活动造成的,而是由于在这些工业交通区中重型卡车的流量较大,也就极大地提高了该地区的噪声水平。)此外还需要对澳门半岛噪声起伏进行研究。对于噪声起伏主要考虑 $L_{max}-L_{eq}$ 和 $L_{eq}-L_{95}$ 两个因子,分别是峰值噪声和本底水平与平均水平的差值出发的,具体数值在表 4 中已经给出。

澳门半岛在昼间和夜间的噪声起伏特征基本上是一致的:

1、 $L_{max}-L_{eq}$ 的峰值噪声水平在绝大部分区域都是超过了 15dB 的国家标准,但基本上又都在 20dB 以下,这说明澳门本身噪声起伏较大,现行的国家标准并不能符合本地区的地理和噪声特点。另外一方面,高噪声复合功能区(混合区、商住交通区、商业交通区和工业交通区)的噪声起伏相对较低,而低噪声复合功能区(休憩区、工业住宅区、交通住宅区和交通

区) 由于本底噪声水平较低, 受瞬间高噪声源的影响较大, 因此其 $L_{max}-L_{eq}$ 要高一些。

2、澳门半岛的 $L_{eq}-L_{95}$ 平均干扰噪声水平都在 10dB 以下, 满足了澳门现行噪声法规定的 A 计权连续等效声级与背景声级之差在 10dB 以下的标准。但不能只用这个指标来判断一个地区的噪声是否超标, 因为这种指标无法反映环境噪声的整体水平, 不利于实现长远的噪声污染控制目标。从复合功能区的角度来看, $L_{eq}-L_{95}$ 的本底噪声水平的基本规律仍然和 $L_{max}-L_{eq}$ 的峰值噪声水平保持一致。该值接近 10dB, 说明已基本没有容量给一些偶发噪声。

3、商业区噪声起伏较小, 居住区起伏较大。这一点是和第 1 点结论基本是一致的, 因为商业区都是在高噪声复合功能区的范围之内, 同时也说明商业区人们频繁的社会活动使得区域噪声水平相当高, 造成偶发噪声对噪声起伏很小; 另一方面, 在住宅区噪声水平较低, 偶发的强噪声源会造成瞬间噪声水平有很大的升高, 是引起居住区噪声起伏较大的原因。

6. 环境交通噪声的控制措施

由于澳门地区的客观环境导致交通噪声水平高, 所以澳门在目前相应的环境噪声标准与中国国家标准相比应该适当放宽, 待以后噪声控制工作进一步加强后再加严。因此在推荐标准的时候, 我们以国标为基础, 适当地参考日本交通环境噪声标准, 将标准的实施分为了两个阶段, 第一阶段的标准相对放宽, 着眼于澳门的现状, 而第二阶段的标准就向国际通用的标准水平靠拢, 这是以后澳门地区控制环境噪声努力的方向。推荐标准如表 5。

表 5 澳门地区推荐环境噪声标准 (单位: dB)

	第一阶段		第二阶段	
	昼间	夜间	昼间	夜间
一类地区	75	60	70	55
二类地区	75	65	75	60

- 注: 1. 一类地区指住宅、学校或医院建筑物外墙 1.5 米以内的区域。
2. 二类地区指一类地区以外的其他区域。
3. 第一阶段夜间最大声级不超过上述数值 20dB, 第二阶段不超过 15dB。
4. 室内噪声标准比相应的室外数值(表中数值)降低 10dB。

7. 结论

研究发现, 澳门半岛交通噪声的影响因素很多, 其中交通流量和车辆行驶速度起着主要作用, 其他一些因素也是不容忽视的, 比如道路坡度、道路封闭程度、地面植被、道路覆盖物等等。这些主要的因素在 STAMSON 模型中都有考虑, 通过增加道路封闭程度修正因子一项, 使 STAMSON 模型达到了较高的精度, 在本研究中最大误差不超过 1.5dB。

在本次研究中, 还利用了地理信息系统 (Mapinfo) 与 STAMSON 模型结果相结合, 建立了用于澳门半岛地区噪声预测与规划的应用系统, 为交通噪声预测与评价提供了技术支持。模拟评估结果表明, 澳门半岛交通噪声水平很高, 夜间持续时间很长, 对整体环境噪声影响很大。此外根据不同功能区监测的噪声水平, 分析了主要噪声源对不同区域的环境噪声影响, 结果表明, 几乎所有区域都不同程度地受到交通噪声干扰, 在白天尤为突出; 商业噪声则成为夜间局部地方产生明显干扰的主要噪声源。最后本文提出了新的澳门半岛环境噪声标准建议, 作为下一步控制交通噪声的法律手段。

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Research of noise simulation and control in Macao

Bai Songtao, Fu Lixin

(Environmental Engineering and Science Department, Tsinghua University)

Wang Zhishi, Tam Vai Man, Chen Haiying, Tang U Wa

(Macao Municipality and Macao University)

Abstract: Traffic noise is the main source of environmental noise in Macao. On the base of the STAMSON model that is popular used in world, according to the character of close road in Macao and building the Close Road Traffic Noise Forecast Model in this article. Through the validation of monitoring data, the result precision of the model is satisfied. Using this model on the traffic noise level in the different type roads in Macao, assessing the quantity pollution level of traffic noise. Finally recommending new Civil Areas Environmental Noise Standard, and suggesting primary measure to amend traffic noise gradually, on the base of noise level on different functional district and the high traffic noise density in Macao.

Keywords: STAMSON, traffic noise, noise simulation functional district

澳门地区饮食业油烟污染特征

马永亮, 张楷, 郝吉明

(清华大学环境科学与工程系, 北京, 100084, 中国)

王志石, 邓宇华

(澳门大学科技学院)

摘要: 本文通过调查和测试, 对澳门地区饮食业单位分布及其油烟污染特征进行研究。分别采用国家环保总局规定方法和 PID 型 VOC 测试仪对 19 家饮食业及食品加工单位的饮食油烟中的油雾和 VOC 同时进行了测定, 对被监测单位基本情况进行调查, 分析了澳门地区饮食业油烟污染的基本情况, 并对澳门地区饮食油烟污染的控制提出建议。澳门地区中小型餐馆数量较多: 所测餐馆平均油烟排放浓度为 $7.2\text{mg}/\text{m}^3$, VOC 的平均浓度为 3.6ppm , 且二者不存在明显相关关系。

关键词: 饮食油烟, VOCs, 空气污染, 澳门

1. 背景

由于中国传统饮食文化的特殊性, 油烟污染问题较为突出。香港、台湾地区以及中国大陆的很多城市在几年前开始对饮食业油烟污染问题进行控制。《中华人民共和国大气污染防治法》中也明确规定, 要求饮食服务业采取有效措施防止油烟对附近居民居住环境的污染, 并规定了行政处罚措施。

2000 年 7 月 1 日国家环保总局开始实施《饮食业油烟排放标准 (试行)》, 确定为强制性国家环境保护标准, 2001 年确定为国标 (GB18483-2001)。标准规定了饮食业单位油烟的最高排放浓度和油烟净化设施的最低去除效率, 并规定排放油烟的饮食业单位必须安装油烟净化设施, 油烟无组织排放视同超标。开始了中国大陆对油烟污染的大规模控制。

澳门地区高楼林立, 街道狭窄, 街区峡谷不利于污染物的扩散。餐饮店遍布各个大街小巷, 镶嵌在民居之间, 油烟对居民生活的影响比较大。澳门地区以博彩和旅游为其特色, 对环境的要求更高。因此, 通过调查和测试对澳门地区饮食业污染的程度和分布特点进行研究, 对于控制油烟污染、改善环境质量具有重要的现实意义。

2. 澳门地区饮食业单位分布特征

据澳门临时市政厅统计, 澳门地区饮食业饭店有 985 个, 按照饭店一次性接待顾客的容量进行分类, 除 119 个饭店没有统计数据外, 容量小于 20 人的饭店为 262 个, 容量为 21-50 的饭店有 435 个, 容量为 51-100 的饭店有 108 个, 容量大于 100 的饭店有 61 个。参见表 1。从饭店数量看, 小型餐馆占据主要部分, 大中型饭店的数量不大, 一次能容纳 100 人以上就餐的饭店仅占 6%左右。顾客与职员比例保持在 5.3 左右。

表- 1 澳门地区饮食业单位统计分布

一次性接待顾客容量 (人)	> 100	51-100	21-50	≤20	无统计 数据	总计
饭店数量, 个	61	108	435	262	119	985
顾客容量总量, 人	15805	7718	14127	4103		41753
职员总数,	3100	1392	2449	916		7857
顾客/职员比率	5.1	5.5	5.8	4.5		5.3

3. 监测方法与调查内容

2001 年 8 月集中对澳门地区的 19 个餐馆排放油烟的情况进行了监测和调查。有鉴于澳门地区饮食业单位规模分布特点，考虑到中国大陆在控制大中型饮食业单位油烟污染控制方面已取得进展，有一定的测试数据和研究基础，澳门地区规模较大的饭店油烟污染状况和控制技术与设备可参考内地的经验。因此，本次旨在摸清饮食业油烟污染排放状况的监测和调查主要集中在中小型餐馆，特别是小型餐馆，餐馆名称及地址参见表-2。所监测调查的餐饮业单位包括了面馆、茶座、大排档、街头小摊、烤肉馆、葡国菜等各种类型，分布于澳门半岛的各个区域。

本次对澳门油烟的监测同时对油雾气溶胶和 VOC 浓度进行测定。对油雾的测定采用《饮食业油烟排放标准》(GB18483-2001)中规定的采样方法和分析方法，即分别采用金属滤筒吸收和红外分光光度法作为测定油烟的采样及分析方法：首先用等速采样法抽取油烟排气筒内的气体，将油烟吸附在油烟雾采集头内；将收集了油烟的采集滤芯置于带盖的聚四氟乙烯套筒中，回实验室后用四氯化碳作溶剂进行超声清洗，移入比色管中定容，用红外分光法测定油烟的含量。烟气中 VOC 的浓度采用 PID 型 VOC 测试仪在进行油烟采样的同时进行测定。

在监测采样的同时调查记录餐馆的基本情况，主要包括餐馆建造时间、每天营业时间、营业面积、从业人数、每天营业收入、所属菜系、每天接待宾客人数、炒菜灶头数量、油烟排放开口位置、每月燃料的使用量等。

表-2 饮食业单位污染源监测及调查的餐馆名称及地址

编号	名称	地址	编号	名称	地址
1	丰顺美食	河边新街	11	饮食广场雄记	
2	康记饭店	河边新街	12	细龟大排档(炒面)	仁慕巷
3	强记大排档	河边新街 65 号	13	珍宝美食	俾利喇街
4	康华美食	木桥街	14	普善斋	罗白沙街
5	侨园美食茶寮	十月初五街	15	澳门街美食	新丽华广场
6	江海美食	黑沙湾马路江海花园	16	河马葡国烧烤	新口岸中土大厦
7	鸭涌河餐厅	孙中山纪念公园	17	喜城海鲜美食	东北大马路
8	馥苑美食	筷子基巴士总站	18	长城烧烤	宋玉生广场
9	金沙餐厅	海景花园	19	九记餐厅	新口岸
10	咀香园				

4. 监测与调查结果分析

4.1. 油烟监测浓度

十九个饮食单位的油烟监测结果见图 1。图中上面一条直线表示十九个点的平均值下面一条线代表中国大陆饮食业油烟排放标准中规定的油烟最高排放浓度限值，分别为 7 mg/m^3 和 2mg/m^3 。从图中可以看出，除个别(两个)饭馆的油烟浓度较高以外，超过 15mg/m^3 ，其余饭馆的油烟平均排放浓度差不多在 5mg/m^3 左右。根据中国大陆制定油烟排放标准时的基础监测数据，大型饭店的油烟初始排放浓度平均为 12mg/m^3 左右，高的可达 50 mg/m^3 以上，中型饭店的初始排放浓度平均为 7mg/m^3 。考虑到本次在澳门地区所测试的饭馆大部分都安装了运水烟罩，该类设备的油烟净化效率约为 30-40%，因此可以推论澳门地区饮食业油烟排放浓度水平与中国内地相当。图 2 为油烟排放浓度的累计分布曲线，由此可见澳门地区不同种类餐馆间的油烟排放浓度不存在显著差异。建议在制定澳门地区饮食业油烟排放标准的浓度限值时借鉴大陆的标准，油烟排放浓度的最高限值取为 2mg/m^3 ，此时如果要求净化装置的净化效率最低应达到 70%以上即可使 95%的中小型餐馆油烟排放满足要求。对于初始排放浓度较高的饭店，其净化装置的效率应相应提高。

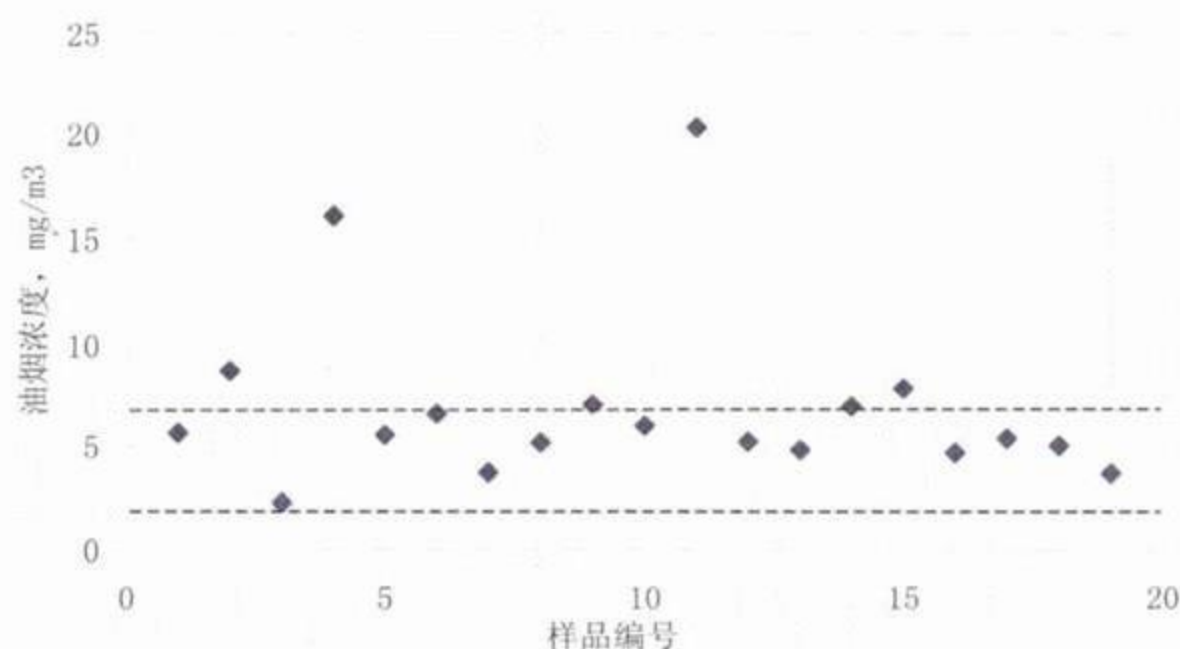


图-1 油烟浓度监测值

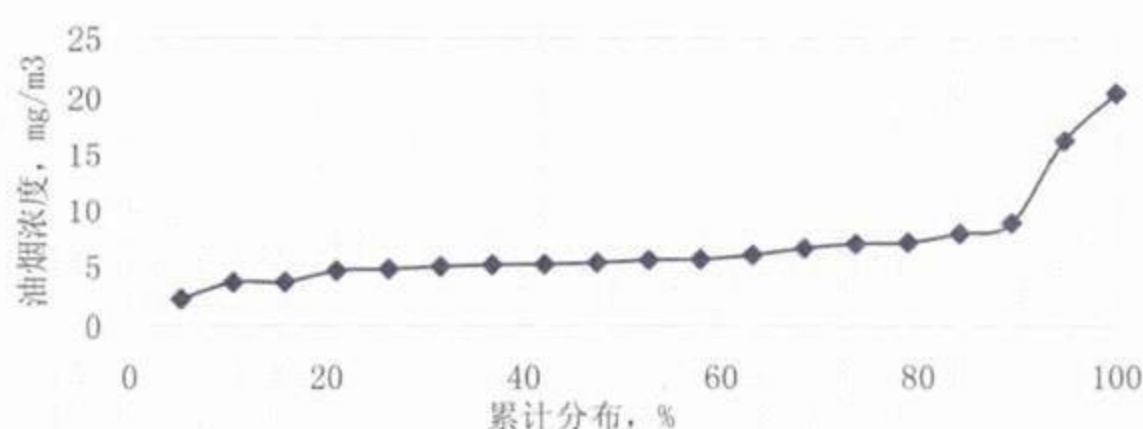


图-2 油烟浓度累计分布图

4.2. 挥发性有机物 (VOC) 浓度测定结果

在进行饭店排放油烟中油雾浓度采样的同时, 采用 PID 型 VOC 仪对油烟中的 VOC 浓度进行同步测定。一方面是要确定油烟中 VOC 的浓度水平, 另一方面是要探讨油烟中油雾浓度和 VOC 浓度的相关性。VOC 浓度的累计分布列于图 3。平均浓度为 3.6ppm, 累计 50% 时的浓度为 3.2ppm, 说明油烟中 VOC 的质量浓度与油雾的浓度基本相当。按照累计 20% 时 VOC 浓度作为油烟排放的控制浓度, 此时 VOC 浓度限值应为 1ppm。如果按照正丁烷分子量进行折算, 也与油烟中油雾的控制浓度相当。

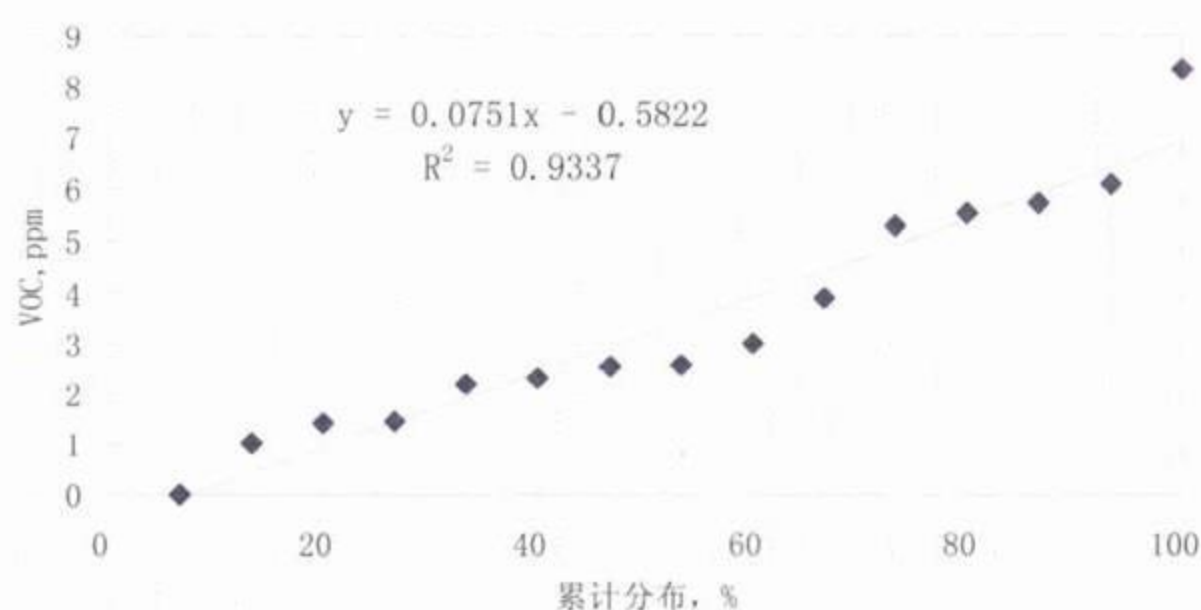


图-3 油烟中 VOC 的浓度累计分布

图 4 为油烟监测中油烟浓度与 VOC 浓度的对应关系图。从图上可以看出, 两者的相关系数的平方值只有 0.011, 从统计学的角度来讲, 不存在相关关系。这与当初要确定 VOC 与油烟浓度的相关关系, 从相关角度出发确定一个监测指标的初衷不相符。因此建议控制饮食业油烟排放污染物的指标应同时包括油烟和 VOC 两项。

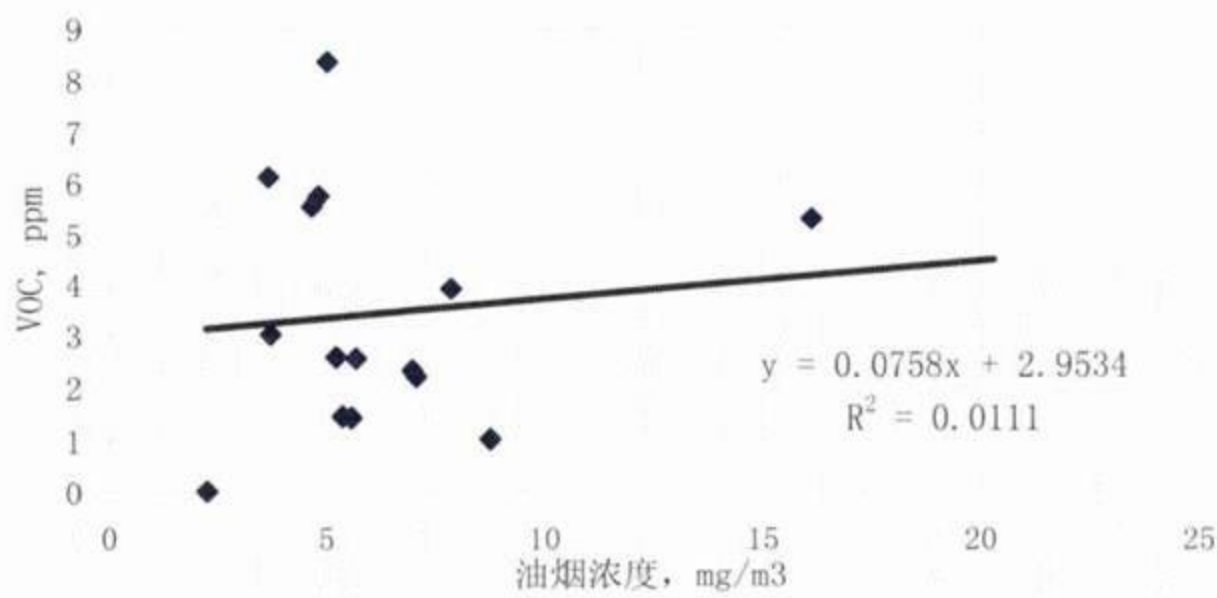


图 4 饮食业排放油烟中油烟浓度与V O C 的相关关系

4.3. 关于燃料问题

在所调查的 19 家餐馆中，所使用的燃料主要包括石油气、煤油（火水）和柴油（油渣），其中有 5 家只使用煤油，每月用量接近 180 桶，折合 2800kg 左右；有 6 家只使用液化气，每月用量 130 罐左右，约折合 2000kg；7 家餐厅同时使用石油气和煤油或柴油，每月消耗石油气 537 罐（8000kg），消耗煤油或柴油 870 桶（11000kg）。总的来看，石油气和煤油的消耗量基本相当，在规模较大的餐馆中煤油的消耗量要大一些。以此推算，澳门饮食业每月消耗的燃料油应在 600 吨以上。石油气是一种清洁的燃料，燃烧过程中不产生异味，而煤油和柴油在燃用过程中均会发出难闻的气味，并有黑烟产生，在采样过程中均能闻到浓浓的柴油味。在中国大陆，大部分的城市地区已经禁止使用柴油和煤油作为饮食业的燃料，全部改用液化石油气或管道天然气或管道煤气作为燃料。只有部分城市的一些饭店还在使用柴油作燃料的灶具，但要求对使用柴油灶具的火烟排放进行治理。鉴于澳门地区的特殊地理环境和政治地位，加之饮食业基本都掺杂在居民聚居地，建议逐渐对煤油和柴油的使用进行限制，直至最后淘汰。

4.4. 澳门地区饮食业油烟污染控制现状

在所调查的餐馆中，大部分的餐馆采用了运水烟罩，少量餐馆安装了静电净化装置，部分餐馆没有任何处理设施。在 19 家餐馆中，采用运水烟罩、静电和没有采取措施的餐馆分别为 10、5、4 家。实际上，运水烟罩的主要作用是调节厨房操作间的温度，改善厨师的操作环境，而对油烟的净化效果有限，只能起到预处理的作用。在香港和中国大陆的南方一些省份，运水烟罩的使用相当普遍，但就油烟污染控制而言，还需同其他设施配合使用。静电技术开始在澳门地区使用，这是一个良好的开端。静电油烟净化器目前已成为大陆控制油烟污染的主力设备，澳门地区应当也会走同样的技术路线。据初步观察，目前澳门地区推广和使用静电油烟净化器还存在着售价过高用户难以承受、选型不配套、维护周期短、净化效率不高等问题。今后可选择推荐大陆近年来发展和筛选出来的一些性能价格比较优的产品，给用户提供更多的选择。

4.5. 其它问题

在饮食广场和街头小摊采样时发现，由于横向气流的影响，炒菜过程中产生的油烟大部分未能吸入排气罩，更谈不上净化的问题了。建议对于露天操作的街头小摊和大排档加以严格限制和管理。

关于建筑时间和开业时间，本次调查中发现，尽管有些建筑的建造时间可以很长，但餐馆的开业时间都不很长，大部分的开业时间在两年以内，最长的也只有五年。因此，饮食业经营的换手率很高。对于饮食业油烟的控制，应充分利用这一特点，规范新开业餐馆的净化设施要求，提高档次，为饮食油烟的有效控制奠定良好基础。

绝大部分餐馆的排气筒都是低空街面排放，在调查中只发现有一例楼顶排放的情况。由于排放高度低，上面一般都由居民楼，因此油烟排放扰民的现象就比较突出。加之澳门高楼

林立, 街道狭窄, 不利于污染物的排放, 油烟污染的危害就更为严重。建议政府鼓励饮食业单位采取措施增加排放高度, 房产开发商在建设过程中即应考虑底层饭馆的排烟问题, 预留通风道等。

5. 结论和建议

1、澳门地区饮食业油烟排放浓度水平与中国内地相当, 调查测试的中小型饮食业单位的油烟平均浓度为 $7.2\text{mg}/\text{m}^3$ 。建议在制定澳门地区饮食业油烟排放标准的浓度限值时最高限值取为 $2\text{mg}/\text{m}^3$, 此时要求净化装置的净化效率最低应达到 70% 以上。对于大型饮食业单位, 器净化装置的效率应达到 90% 以上。

2、监测结果表明, 饮食业油烟中油雾浓度与 VOC 浓度间不存在明显的相关关系, 因此控制饮食业油烟排放污染物的指标应同时包括油烟和 VOC 两项。

3、所测试油烟中油烟中 VOC 的质量浓度与油雾的浓度基本相当。

4、澳门地区饮食业中气体燃料和燃料油的使用量基本相当。煤油和柴油在燃用过程中均会发出难闻的气味, 并有黑烟产生, 污染相对较重。特区政府应逐渐对煤油和柴油的使用进行限制, 用清洁燃料予以替代, 直至最后淘汰。

5、目前澳门地区推广和使用静电油烟净化器还存在着售价过高用户难以承受、选型不配套、维护周期短、净化效率不高等问题。今后可选择近年来大陆发展和筛选出来的一些性能价格比较优的产品推荐给用户, 并建立示范工程, 为其提供更多的选择和更优的服务。

6、对于饮食业油烟的控制, 应充分利用饮食业单位换手率高的特点, 规范新开业餐馆的净化设施要求, 为饮食油烟的有效控制奠定良好基础。

7、应鼓励饮食业单位采取措施增加排放高度, 房产开发商在建设过程中即应考虑底层饭馆的排烟问题, 预留通风道。

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在油烟采样点的选择和样品采集过程中, 澳门临时市政厅的工作人员给予了大力的支持, 特别是陈海英女士, 全程参加本次调研, 并做了大量的准备工作。在此对于给予过我们帮助的有关人士表示衷心感谢。

Characterization of Cooking Fume Pollution in Macao

MA Yongliang, HAO Jiming, ZHANG Kai
(Dept. of Envir. Sci. & Engr., Tsinghua University, Beijing, 100084, China)
WANG Zhishi, Tang U Wa
(Faculty of Technology, Macao University)

Abstract: The characteristics of the size distribution of restaurants in Macao and related air pollution were studied via site monitoring and investigation. The concentrations of oil fume from 19 small/medium sized restaurants or food processing workshops have been measured with the infrared method, and VOCs concentrations in the cooking fume were determined with VOC monitor with PID method during the oil fume sampling. The status of the restaurant was also recorded with a investigation questionnaire. The mean concentrations of oil fume and VOCs were $7.2\text{ mg}/\text{m}^3$ and 3.6ppm respectively, but there was no obvious correlation between them. Some suggestions on pollution control from restaurants in Macao area were presented.

Keywords: cooking fume; VOCs; air pollution; Macao

非规则网格下二维浅水流动的数值模拟*

陈祖华 王光谦

(清华大学教育部水沙科学重点实验室, 北京 100084)

王志石

(澳门大学科技学院, 澳门)

摘要: 本文建立了一种在非规则结构化网格上求解平面二维浅水流动的有限体积方法, 通过采用地形在离散网格内双线性变化及离散网格界面间地形连续的地形逼近方法, 应用可以有效处理间断问题的 Roe 格式来离散浅水方程中的对流项, 并通过 Van Leer 提出的状态插值法提高格式精度。在计算原始变量在网格内的插值梯度时, 采用最小二乘方法求变量的最优梯度代替差分计算梯度, 从而可采用任意形状的不规则四边形网格离散计算域。计算实例表明, 本文的方法能够计算间断问题并能够处理各种复杂流态的过渡, 具有较好适应性和计算精度, 能够满足不同实际问题的计算要求。

关键词: 浅水方程 有限体积法 Roe 格式 潮流 澳门周围水域

1. 引言

许多工程中的流动问题常常可以用浅水流动模型来描述, 如近海、湖泊、水库及河口中的流动。由于应用广泛, 对浅水流动控制方程组的数值方法也受到较多关注。由于浅水方程在数学上属双曲方程组, 在一定的条件下会产生如水跃、溃坝波、涌浪等的强间断解, 这给方程的数值求解带来了一些困难, 普通的有限差分格式不能有效处理间断解, 因而大大限制了所编程序的应用范围。近年来, 在计算空气动力学中已经较为成熟的许多高精度无振荡的激波捕捉格式, 如 TVD 格式、利用黎曼解的 Godunov 型格式等, 这些格式克服了间断产生的数值振荡, 可以处理复杂流态的过渡, 正越来越多地被应用到浅水流动计算中, 如[1]~[3]。与气流不同, 由于浅水流动存在底部地形的变化, 当地形变化较大时不能直接应用这些格式。

本文通过采用地形在离散网格内双线性变化及离散网格界面间地形连续的地形逼近方法, 采用可以有效处理间断问题的 Roe 格式^[5]来离散浅水方程中的对流项, 在此基础上建立了一种平面二维浅水方程的数值求解方法。采用 Van Leer 提出的状态插值法 (MUSCL)^[6]提高格式精度, 界面状态插值时采用了文献[1]中使用的方法即以水位插值代替水深插值, 以解决在复杂地形条件下应用 Roe 格式的问题。在计算原始变量在网格内的插值梯度时, 本文采用最小二乘方法求变量的最优梯度代替差分计算梯度, 从而可采用任意形状的不规则四边形网格离散计算域, 这避免采用正交曲线坐标变换方法以拟合很多实际问题中计算域的不规则的边界, 降低了网格生成的困难, 同时又能具有较高的边界拟合精度。

2. 基本方程

采用如下守恒形式的水深平均平面二维流动控制方程组:

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作者简介: 陈祖华 (1974—), 男, 贵州省毕节市人, 清华大学水利系水沙科学重点实验室博士研究生, 主要从事河口海岸泥沙数学模型研究。

$$\frac{\partial \mathbf{U}}{\partial t} + \frac{\partial \mathbf{F}}{\partial x} + \frac{\partial \mathbf{G}}{\partial y} = \frac{\partial \tilde{\mathbf{F}}}{\partial x} + \frac{\partial \tilde{\mathbf{G}}}{\partial y} + \mathbf{b} \quad (1)$$

$$\mathbf{U} = \begin{bmatrix} h \\ hu \\ hv \end{bmatrix} \quad \mathbf{F} = \begin{bmatrix} hu \\ hu^2 + \frac{1}{2}gh^2 \\ huv \end{bmatrix} \quad \mathbf{G} = \begin{bmatrix} hv \\ huv \\ hv^2 + \frac{1}{2}gh^2 \end{bmatrix} \quad \tilde{\mathbf{F}} = \begin{bmatrix} 0 \\ \tau_{xx} \\ \tau_{yx} \end{bmatrix} \quad \tilde{\mathbf{G}} = \begin{bmatrix} 0 \\ \tau_{xy} \\ \tau_{yy} \end{bmatrix} \quad (2)$$

$$\mathbf{b} = \begin{bmatrix} 0 \\ gh(S_{0x} - S_{fx}) + hfv \\ gh(S_{0y} - S_{fy}) - hf u \end{bmatrix} \quad (3)$$

式中, u, v 分别为 x, y 方向的水深平均流速分量, h 为水深, g 为重力加速度, ρ 是水的密度。 $\tau_{xx}, \tau_{xy}, \tau_{yx}$ 和 τ_{yy} 为:

$$\tau_{xx} = 2h\nu_t \frac{\partial u}{\partial x}, \quad \tau_{xy} = \tau_{yx} = h\nu_t \left(\frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} \right), \quad \tau_{yy} = 2h\nu_t \frac{\partial v}{\partial y} \quad (4)$$

其中 ν_t 是紊动粘性系数, 在一般的计算中可以采用常数或简单的零方程模型计算。

式 (3) 中 S_{0x} 和 S_{0y} 分别表示 x 和 y 方向的底坡。 f 为 Coriolis 系数, S_{fx}, S_{fy} 是底部摩阻坡降,

$$S_{fx} = \frac{C_f \sqrt{u^2 + v^2}}{gh} u, \quad S_{fy} = \frac{C_f \sqrt{u^2 + v^2}}{gh} v \quad (5)$$

其中 C_f 为底部阻力系数, 采用常用的曼宁公式则有 $C_f = g \frac{n^2}{h^{1/3}}$, 这里 n 是曼宁系数。

3. 3. 数值求解方法

本文采用有限体积法求解方程 (1)。有限体积法的关键在于计算对流通量的格式。考虑到浅水方程双曲性和非线性特点, 本采用一种有效处理间断问题的 Roe 通量差分裂格式^[5], 该格式具有自动处理复杂流态过渡, 无振荡等优点, 因此本文所用的数值方法不仅适用与一般的光滑流动, 也能计算溃坝水流或有水跃等复杂流态过渡的流动问题。采用这种无振荡的格式也利于在潮汐流动计算中处理活动边界。由于这种利用黎曼解的格式是由计算空气动力学推广而来, 因此必须要考虑浅水流动有别于气流的方面^[7], 主要是在浅水流动中要考虑水下地形的变化。

为了在水下地形变化较大时也能够直接推广应用 Roe 格式, 本文采用任意四边形网格离散计算域, 并假定每一网格内的地面高程双线性变化, 网格间的交界面上底高程保持连续, 如图 1 所示, 这时一个离散网格内的地形分布由其四个顶点的高程决定。

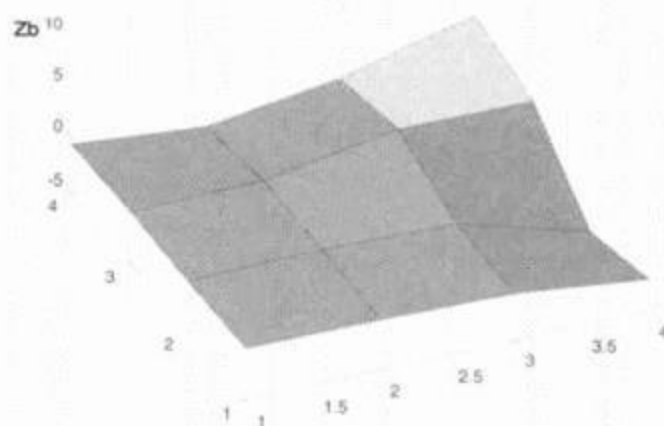


图 1 地形逼近示意
Fig.1 Sketch of Approximation to Topography

采用有限体积法离散方程 (1), 如图 2 所示, 记 Ω_c 为网格的面积, 时间离散采用一阶显格式, 得到:

$$\Omega_c \frac{U^{n+1} - U^n}{\Delta t} + \sum_j F_{\perp j}(U^n, n_j) l_j = \sum_j \tilde{F}_{\perp j}(U^n, n_j) l_j + b(U^n) \quad (6)$$

式中 Δt 是时间步长, l_j 表示网格的第 j 边的边长, $F_{\perp j}$ 和 $\tilde{F}_{\perp j}$ 分别是网格第 j 边的法向对流和扩散项数值通量, 以网格边中点处的值代表, n_j 表示网格的第 j 边的外法向向量。对扩散通量, 采用中心格式离散, 即

$$\tilde{F}_{\perp}(U, n) = \tilde{F}(\bar{U}) \cdot n_x + \tilde{G}(\bar{U}) \cdot n_y \quad (7)$$

式中 n_x 和 n_y 是外法向 n 的两个分量, $\bar{U}$ 表示变量在网格界面处的平均值。

对流项数值通量的计算格式对浅水方程的数值求解很重要。本文采用 Roe 通量差分裂格式^[5], 它是一种基于近似黎曼解的 Godnov 型格式。 $F_{\perp}$ 可写作:

$$F_{\perp}(U, n) = T^{-1} \Phi^{Roe}(V_L, V_R) \quad (8)$$

式中 T 是网格界面处的局部坐标变换矩阵,

$$T = \begin{bmatrix} 1 & 0 & 0 \\ 0 & n_x & n_y \\ 0 & -n_y & n_x \end{bmatrix} \quad T^{-1} = \begin{bmatrix} 1 & 0 & 0 \\ 0 & n_x & -n_y \\ 0 & n_y & n_x \end{bmatrix} \quad (9)$$

V 是局部坐标系中的守恒变量向量, $V = TU = [h \ hU_{\perp} \ hU_{\parallel}]^T$, 其中 $U_{\perp}$ 表示网格边外法向法向流速分量而 $U_{\parallel}$ 表示平行与网格边方向的流速分量。下标 L, R 表示网格两侧的状态。 $\Phi^{Roe}(V_L, V_R)$ 是 Roe 的一阶迎风格式数值通量,

$$\Phi^{Roe}(V_L, V_R) = \frac{1}{2} [F(V_L) + F(V_R) - \tilde{R} |\tilde{\Lambda}| \Delta \tilde{V}] \quad (10)$$

式中, $|\tilde{\Lambda}|$ 是一个对角阵, 其对角线上的元素是通量向量 F 的 Jacobi 矩阵的特征值取绝对值,

$$|\tilde{\Lambda}| = \begin{bmatrix} |\tilde{U}_{\perp} - \tilde{a}| & 0 & 0 \\ 0 & |\tilde{U}_{\perp}| & 0 \\ 0 & 0 & |\tilde{U}_{\perp} + \tilde{a}| \end{bmatrix} \quad (11)$$

其中 $\tilde{a} = \sqrt{gh}$ 。矩阵 R 的每一列是 F 的 Jacobi 矩阵的右特征向量

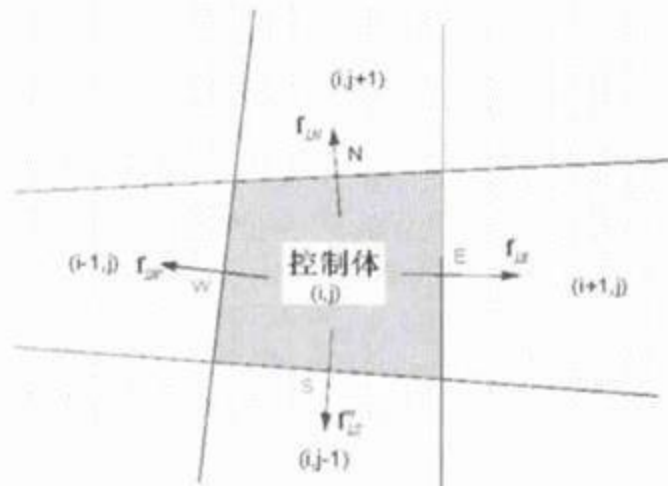


图 2 有限体积离散示意
Fig.2 Sketch of Typical Cell

$$\tilde{\mathbf{R}} = \begin{bmatrix} 1 & 0 & 1 \\ \tilde{U}_\perp - \tilde{a} & 0 & \tilde{U}_\perp + \tilde{a} \\ \tilde{U}_\parallel & 1 & \tilde{U}_\parallel \end{bmatrix} \quad (12)$$

$\Delta \mathbf{V} = \mathbf{R}^{-1}(\mathbf{V}_R - \mathbf{V}_L)$, 它表示跨网格边界的守恒变量差的特征强度,

$$\Delta \tilde{\mathbf{V}} = \begin{bmatrix} \frac{\tilde{U}_\perp + \tilde{a}}{2\tilde{a}}(h_R - h_L) - \frac{1}{2\tilde{a}}(h_R U_{\perp R} - h_L U_{\perp L}) \\ -\tilde{U}_\parallel(h_R - h_L) + (h_R U_{\parallel R} - h_L U_{\parallel L}) \\ -\frac{\tilde{U}_\perp - \tilde{a}}{2\tilde{a}}(h_R - h_L) + \frac{1}{2\tilde{a}}(h_R U_{\perp R} - h_L U_{\perp L}) \end{bmatrix} \quad (13)$$

式中变量上标‘~’表示取 Roe 平均值, 对二维浅水方程, 其表达式如下,

$$\tilde{h} = \frac{1}{2}(h_L + h_R), \quad \tilde{U}_\perp = \frac{\sqrt{h_L} U_{\perp L} + \sqrt{h_R} U_{\perp R}}{\sqrt{h_L} + \sqrt{h_R}} \quad (14)$$

$\tilde{U}_\parallel$ 的公式与 $\tilde{U}_\perp$ 类似。Roe 格式不能识别稀疏波与膨胀间断, 因此在遇到零特征值时会产生不正确的间断解, 因此要作修正, 本文采用了 Monthe L.A. et al (1999) 文中的方法, 修正后的 Roe 格式是具有正性的。

Roe 格式只有一阶精度, Van Leer (1979) 提出的状态插值法(MUSCL)^[6]是将 Godnov 型格式改进到二阶精度的一种常用方法, 记 $\mathbf{W} = [h \ u \ v]^T$ 为原始变量向量, 对平面二维情形, 在网格编号 i 变化方向变量插值采用下式

$$\mathbf{W}_{i+\frac{1}{2},j}^L = \mathbf{W}_{i,j} + \frac{\partial \mathbf{W}}{\partial x} \Big|_{i,j}^{\lim} (x_{i,j}^E - x_{i,j}^G) + \frac{\partial \mathbf{W}}{\partial y} \Big|_{i,j}^{\lim} (y_{i,j}^E - y_{i,j}^G) \quad (15a)$$

$$\mathbf{W}_{i+\frac{1}{2},j}^R = \mathbf{W}_{i+1,j} + \frac{\partial \mathbf{W}}{\partial x} \Big|_{i+1,j}^{\lim} (x_{i,j}^E - x_{i+1,j}^G) + \frac{\partial \mathbf{W}}{\partial y} \Big|_{i+1,j}^{\lim} (y_{i,j}^E - y_{i+1,j}^G) \quad (15b)$$

式中 $\frac{\partial \mathbf{W}}{\partial x} \Big|_{i,j}^{\lim}$ 和 $\frac{\partial \mathbf{W}}{\partial y} \Big|_{i,j}^{\lim}$ 是 $\mathbf{W}$ 在网格内的受限制的插值坡度在 x 和 y 方向的分量, x 和 y 表示

坐标, 上标 E 表示网格边界中点, G 表示网格形心 (参见图 2), 沿编号 j 方向的插值公式与此类似。对非规则的四边形网格, 不能简单地通过差分得到原始变量的变化坡度。本文按如下方法确定变化坡度: 对每个原始变量, 其变化坡度在 x 和 y 方向的分量是如下二元平方函数的极小值点:

$$\Psi\left(\frac{\partial f}{\partial x}, \frac{\partial f}{\partial y}\right) = \sum_{p \in K(i,j)} \left| (f_{i,j} - f_p) + \frac{\partial f}{\partial x} (x_p^G - x_{i,j}^G) + \frac{\partial f}{\partial y} (y_p^G - y_{i,j}^G) \right|^2 \quad (16)$$

式中 $K(i,j)$ 表示与待求网格具有相邻边的网格的集合。方程(17)可以用最小二乘方法求解。为了防止插值引起的数值振荡, 要对原始变量在网格内的插值梯度进行限制, 这由下面的限制器函数得到:

$$\frac{\partial f}{\partial x} \Big|_{i,j}^{\lim} = \frac{1}{2} \left[\min_{p \in \hat{K}(i,j)} \operatorname{sgn}\left(\frac{\partial f_p}{\partial x}\right) + \max_{p \in \hat{K}(i,j)} \operatorname{sgn}\left(\frac{\partial f_p}{\partial x}\right) \right] \min_{p \in K(i,j)} \left(\left| \frac{\partial f_{i,j}}{\partial x} \right|, 2 \left| \frac{\partial f_p}{\partial x} \right| \right) \quad (17)$$

式中 $\hat{K}(i, j) = K(i, j) Y \{(i, j)\} \cdot \frac{\partial f}{\partial y} \Big|_{i,j}^{\text{lim}}$ 采用同样的方法得到。采用这种方法而不是使用

单侧差分求变量的变化梯度, 虽然计算量和格式数值粘性均有所增加, 但是可以采用不规则网格, 从而降低了网格生成的困难, 又能提高不规则边界的拟合精度。

但是对于浅水流动来说, 由于存在水下地形的变化, 有时地形变化还较大, 这时采用以上的插值方法不适用, 一般的处理方法是将水深插值代以水位插值, 在插值出网格界面中点水位后, 减去该点水底高程得到水深^[1]。这也是为什么水下地形的描述方式要求保证网格界面地形连续的原因。

4. 边界条件及动边界处理

采用虚网格法来设置边界条件, 通过把边界条件加在对应的虚网格上, 这样可以使边界通量的计算采用与域内相同的方法计算, 使边界格式与内点一致。边界条件可分为固壁边界和开边界两种。对固壁边界, 法向流速为零而切向常采用滑移条件。其相应的虚网格的变量值采用镜像条件确定。即具有与内点相同的水深而流速是内点流速关于边界的镜像, 同时其状态插值的受限坡度 δ 与内点在边界法向投影大小相等, 方向相反^[2]。对开边界虚网格上的未知量, 本文采用变量外插来确定。

动边界问题常常是河口海岸地区数值模拟中需要考虑的, 它也是数值计算中一个比较困难的问题。采用 Roe 格式有利于处理动边界问题, 因为经过修正后的 Roe 格式已满足熵条件且具正性, 能够正确计算有水网格与无水网格界面的数值通量 (它实际上是一个下游无水时的溃坝问题), 不会因数值振荡引起负水深导致计算无法进行, 无需特别的处理。但是对网格取平均后在动边界处常常具有偏高的流速, 为了保证计算稳定, 仍然需要对动边界作一定的处理。本文采用的方法是将两侧网格都是不完全淹没网格的网格边界当作固壁边界处理以阻止动边界处的水深扩散, 避免较高的流速。这里所谓不完全淹没指网格形心的水位小于任意一个网格节点的水下高程。这可以看作是小水深法的一种变体, 但是它更好地保持了有限体积法的守恒优点。此外, 在动边界附近, 为了防止摩阻项引起计算失稳, 需要对该项作一些限制, 限于篇幅不在此处讨论。

5. 计算实例与讨论

本文采用两个模型问题和一个实际算例来检验所提出的方法。第一个例子是一维溃坝水流的水槽实验, 它检验数值格式处理间断、复杂流态过渡和动边界的能力; 第二个例子是计算 Bellos 等^[9]的二维溃坝水流的水槽实验, 检验本文方法处理二维间断水流计算的效果; 澳门周围水域潮流计算说明本文方法在工程实例中的应用效果。

5.1. 一维溃坝水流计算

这个例子取自文献[4]。等宽水槽中设置一个截面为三角形的堰, 在距左端 15.5m 处设有闸门, 闸门上游水深 0.75m (图 4 中虚线), 闸门与堰之间无水。水槽末端为高 0.15m 的挡水堰, 它与上述三角形堰之间保持 0.15m 深的静水。在 $t=0$ 时突然将闸门移去。计算采用均匀网格, 空间步长 0.5m, 计算中对下游边界作了简化, 没有考虑末端挡水堰而是将下游看作静水深 0.15m 的无限长渠道。计算中忽略扩散项, 曼宁糙率取 0.0125^[4]。图 3 是距水槽左端 19.5m 处的水位随时间的变化过程计算值与实验量测值的比较, 二者符合较好。图 4 中实线是 $t=10s$ 时水位沿程分布, 可以看到在三角形堰前后水流均发生了水跃。除下游段因边界简化而与文献[4]中有所差异外, 本文计算的结果与原文献是一致的。

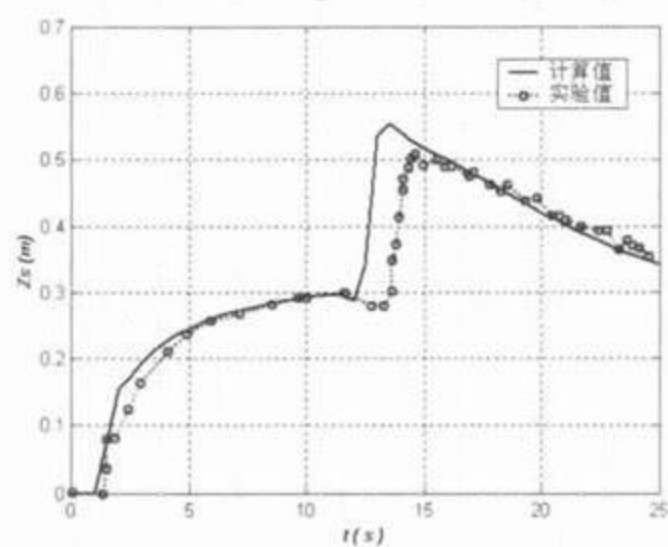


图 3 x=19.5m 处水位变化过程
Fig.3 Water level at x=19.5m

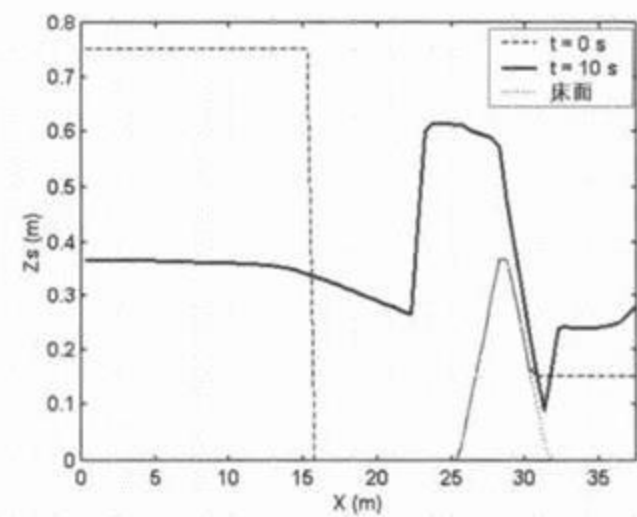


图 4 t=10s 时水面线的沿程分布的计算结果
Fig.4 The computed profile of water surface at t=10s

5.2. 二维溃坝水流

这个例子是 Bellos 等^[9]进行的一个瞬时溃坝水流的水槽实验。实验水槽如图 5 所示。水槽中部有一个收缩-扩散段，闸门位于其中最窄处，瞬时打开。所选的计算条件是上游水深 30cm，下游水深 10.1cm，底坡为 0。计算糙率取 0.012，共划分为 42×10 个网格，下游末端的边界条件为矩形薄壁堰的水位流量关系。图 6 是几个测点的水位过程线的计算和实测值的对比，其中 x=-0 指上游紧靠闸门处的测点。由图可见，各测点的水位过程线的计算值均很好地和实测值符合。

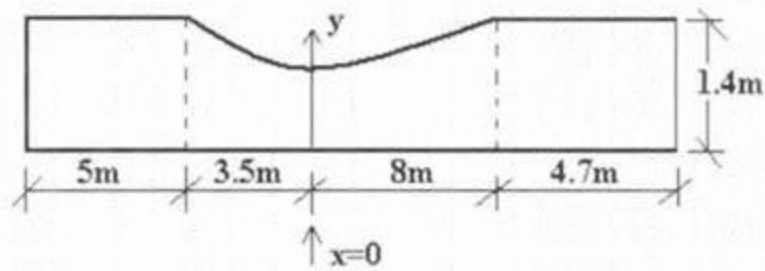
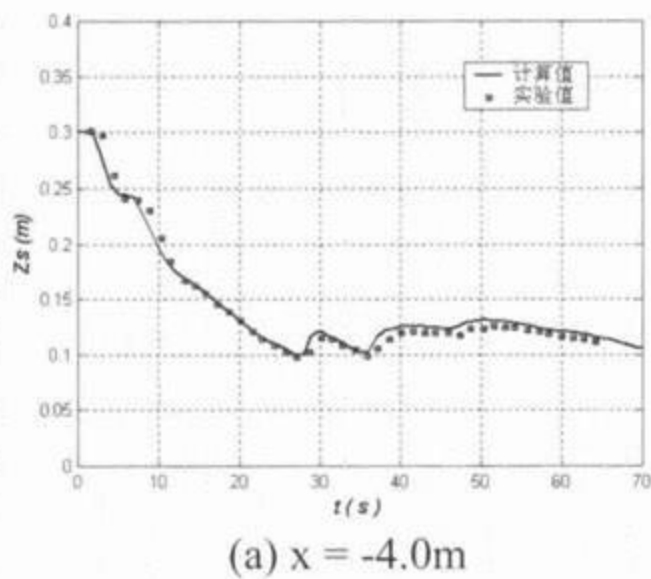
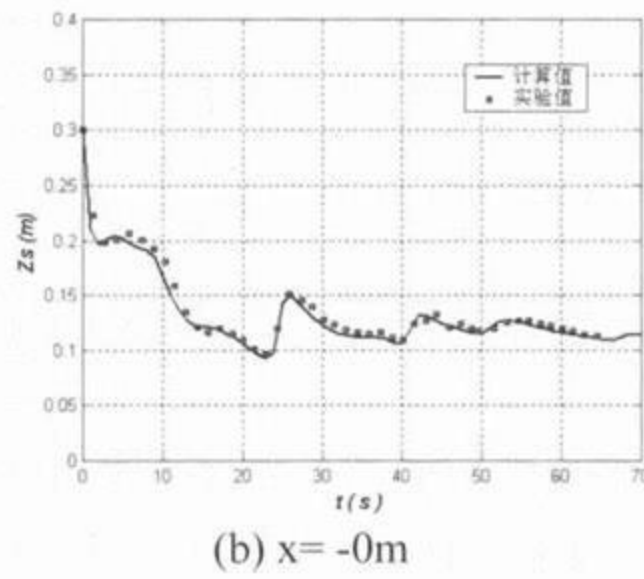


图 5 Bellos 等的实验水槽平面形状
Fig.5 The plan of flume in Bellos et al.'s experiment



(a) x = -4.0m



(b) x = -0m

图 6 不同测点的水位变化过程线
Fig.6 Hydrographs of water levels in two points

5.3. 澳门周围水域潮汐流动的计算

澳门附近水域东临伶仃洋湾口之西侧、往西连接磨刀门水道的支汊洪湾水道；南面是珠江口外海区。在上游径流、外海潮流、沿岸流、风浪流等水动力共同作用影响下，水流运动较为复杂。我们采用上述数值方法对澳门周围水域的潮汐流动进行了计算，为了利用现有资料，计算采用均匀矩形网格，计算区域为 32.0km×22.5km 的矩形（图 7），共划分为 65×46 个网格，网格大小为 500m×500m。

采用 1980 年 11 月的一组水文资料^①对澳门周围水域的潮汐流动作了验证计算, 计算中糙率取 0.017。边界条件的确定方法如下: 对南边界采用水位边界条件, 由实测潮汐水位指定。对西江入口处给定流量, 由于资料有限, 只能给定一个恒定流量, 忽略了时间变化过程。其余开边界无资料给定, 采用人工数值边界条件。

图 8 是计算的 P4 点水位与实测资料的对比。由图可见涨落潮的潮位、相位和波形均与实测吻合良好, 计算基本反映了该地区的潮位变化规律。图 9 是 P3 测点的流速计算值与实测值的对比, 两者较为符合, 只是开始几小时流向误差较大, 这一方面可能是因为在平潮时流速较小, 可能的测量误差较大, 另一方面是也可能是因为数值边界条件未能完全反映来自伶仃洋的潮流的影响造成的误差。

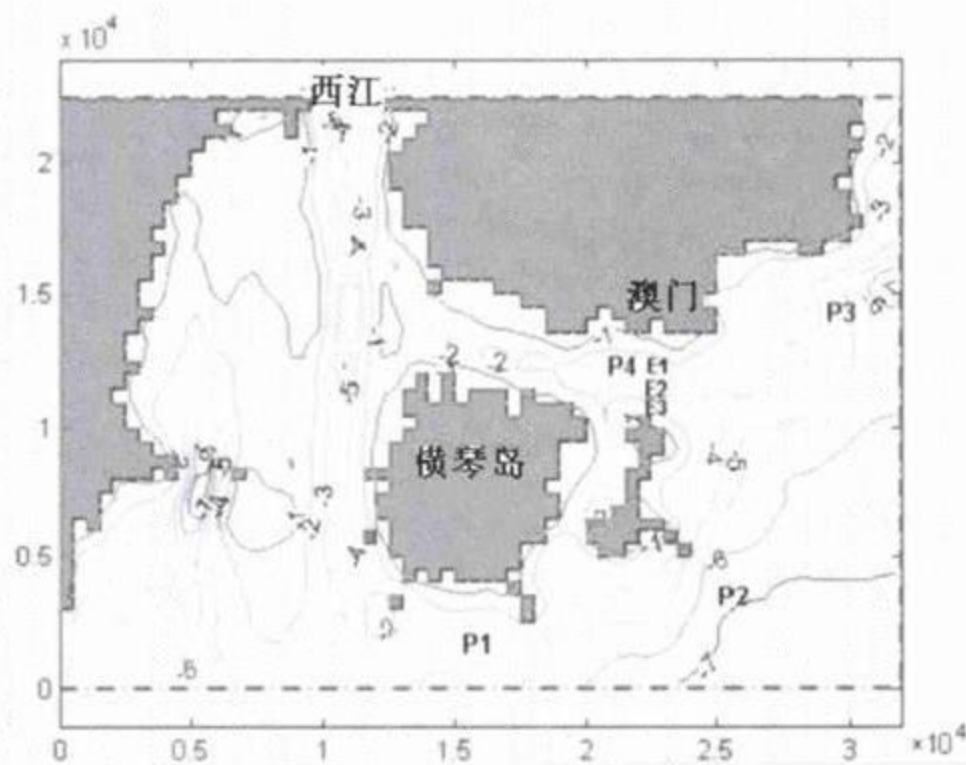


图 7 计算区域地形等高线

Fig.7 Computational Domain

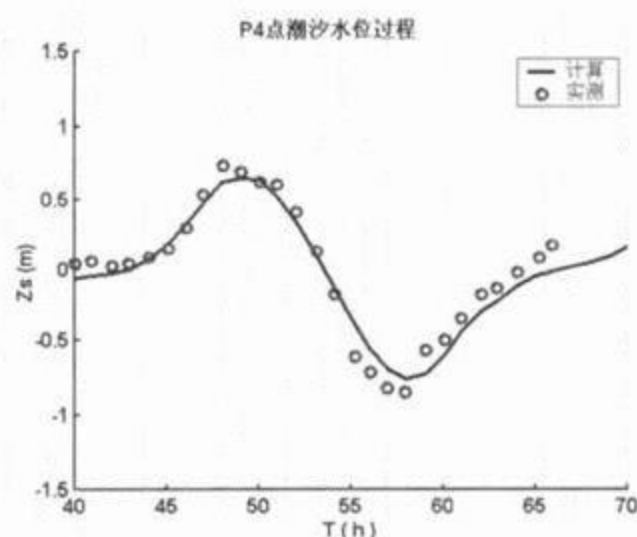


图 8 P4 点水位过程计算与实测对比

Fig.8 Water level at Station P4

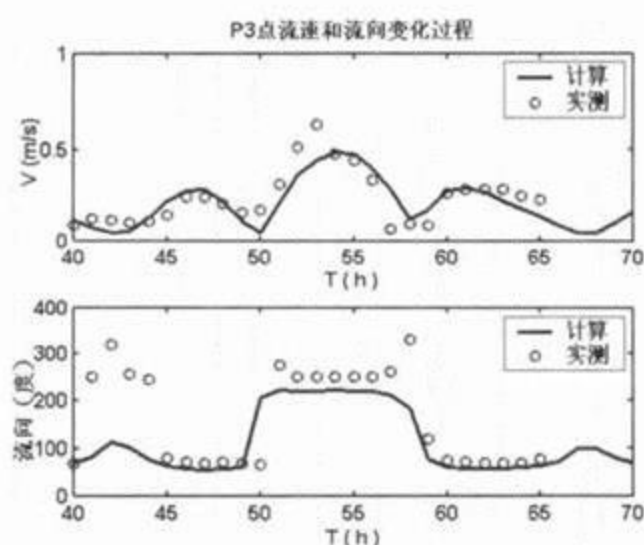


图 9 P3 点流速流向验证

Fig.9 Velocity and direction at Station P3

6. 结语

在浅水流动计算中非线性对流项的数值离散格式对计算是非常重要的, 本文将基于黎曼解的 Roe 格式结合 MUSCL 方法应用平面二维浅水流动的数值计算中, 在此基础上建立了一种数值求解平面二维浅水流动的数值方法。由于采用最小二乘方法解网格内的最优梯度代替单侧差分, 本文方法的人工数值耗散较一般 TVD 格式大, 但是可以直接应用于任意不规则的离散网格上而不采用坐标变换, 在降低网格剖分要求的同时增加了对物理边界拟合的精度。计算实例表明, 由于采用了性能较好的 Roe 格式, 本文的方法能够计算间断问题并能够处理各种复杂流态的过渡, 具有较好适应性和计算精度, 能够满足不同实际问题的计算要求。

^① Instituto Hidrográfico (1992) The numerical hydromorphological model of the surrounding waters of Macao, Final report.

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Numerical Simulation of Two-Dimensional Shallow Water Flow on Irregular Grids

Chen Zuhua Wang Guangqian

(Tsinghua University, Key Laboratory for Water and Sediment sciences of Ministry of Education,
Beijing 100084)

Wang Zhishi

(The College of Science and Technology, University of Macau)

Abstract: A numerical model for the solution of the two-dimensional shallow-water flow on irregular grids is described. The model, which is based on finite-volume method, adopted the Roe's approximate Riemann solver to deal with the advection terms in the shallow-water equations. A bilinear approximation for the topography in a discrete cell is introduced to preclude the restrictions on the Roe's scheme due to bed slope. And a modified MUSCL technique is combined to obtain an increased accuracy. Instead of unilateral difference method, the least square method is used to find the gradient of the primitive variables so that irregular grids can be used. The results of several examples show that the model is capable of treating discontinuities and flow transients, and is of good accuracy as well as performance for different applications.

Key words: Shallow-Water Equations, Finite-Volume Method, Roe's scheme, Tidal Flow, The Surrounding waters of Macau

河口抛泥数学模型及应用

陈祖华 王光谦

(清华大学教育部水沙科学重点实验室, 北京 100084)

王志石

(澳门大学科技学院, 澳门)

摘要: 在平面二维潮流数学模型的基础上, 采用扩散模型建立了河口区抛泥引起的泥沙对流扩散数学模型, 并利用该模型对澳门周围水域疏浚土水抛后的泥沙迁移过程进行了模拟, 对澳门水域抛泥后泥沙的迁移作了初步分析。

关键词: 数学模型; 抛泥; 扩散; 澳门水域

在港口工程中港池和航道的开挖以及维护航道进行的疏浚活动都要涉及到对疏浚泥土的处理, 根据各地具体条件, 可以采用分别陆抛和水抛等两种常见的方法处理。疏浚土水抛后, 一部分泥沙沉积在抛泥点附近, 其余泥沙会在抛泥点局部区域形成高浓度含沙水体, 并随潮汐水流运动, 有时运动到人们不希望的区域, 因此, 研究疏浚土水抛以后在水体中的输移情况对分析这种人为活动所造成的对生态环境的影响是非常重要的。

疏浚土进入水体后的初期运动比较复杂, 对抛泥初始阶段的泥沙运动机理的研究尚不充分, 但是根据抛泥的现场调查发现, 疏浚土进入水体后, 大部分泥沙将快速沉入抛泥点附近水底, 另一部分则随潮汐水流对流扩散输移, 随着泥沙浓度减低, 泥沙在水中的运动与一般水流中泥沙的对流扩散运动并无分别。如果不特别关心抛泥近区运动的细节, 只要对抛泥初始阶段作出合理的概化, 仍然可以采用工程上常用的平面二维泥沙数学模型对抛泥后期疏浚土随潮流迁移扩散趋势作出一定程度的预测, 以评价抛泥对工程水域的影响。本文采用瞬时点源扩散模型建立了一种抛泥远区数值模拟方法, 并以澳门水域抛泥为例, 对抛泥引起的泥沙迁移情况作了数值模拟研究。

1. 河口区平面二维泥沙运动数学模型

1.1. 基本方程

描述悬浮泥沙在水中对流扩散的平面二维方程如下:

$$\frac{\partial hC}{\partial t} + \frac{\partial huC}{\partial x} + \frac{\partial hvC}{\partial y} = \frac{\partial}{\partial x} \left(hD_x \frac{\partial C}{\partial x} \right) + \frac{\partial}{\partial y} \left(hD_y \frac{\partial C}{\partial y} \right) + Sc \quad (1)$$

式中 C 表示泥沙的水深平均浓度, D_x 和 D_y 泥沙的混合扩散系数 (紊动扩散系数加离散系数)。 Sc 为源项, 具有如下形式:

$$Sc = E - D \quad (2)$$

E 表示泥沙的冲刷通量, D 为泥沙的沉降通量。 E 采用下式计算 (Partheniades, 1965):

$$E = M_E \max\left(0, \frac{\tau_b}{\tau_{ce}} - 1\right) \quad (3)$$

式中 M_E 为冲刷率常数, τ_b 是床面切应力, τ_{ce} 为临界冲刷切应力。

沉降通量 D 的计算公式为 (Krone, 1962):

$$D = pC_b W_{sb} \quad (4)$$

式中 C_b 表示悬沙近底浓度, W_{sb} 为相应的沉速, p 为悬沙沉降几率,

$$p = \max(0, 1.0 - \frac{\tau_b}{\tau_{cd}}) \quad (5)$$

式中 τ_{cd} 是临界沉降切应力。悬沙近底浓度的确定方法是假定 $C_b = \beta C$, 其中 β 按下式计算 (Metha et al., 1989):

$$\beta = 1 + \frac{Pe}{1.25 + 4.75p^{2.5}} \quad (6)$$

式中 Pe 为悬沙沉降 Peclet 数, $Pe = \frac{\sigma_c W_s h}{\nu_t}$ 。

泥沙沉降速度 W_s 的计算方法为: 当悬沙浓度较低时 ($C < C_{free} = 0.1 \sim 0.3 \text{ g/L}$), 絮团间的相互影响不显著, 处于自由沉降阶段, 这时的沉速可按 Stokes 公式计算:

$$W_s = \frac{(s_f - 1)gd_m^2}{18\nu} \quad (C < C_{free}) \quad (7)$$

式中 s_f 是悬浮絮团的比重, d_m 为絮团中值粒径, ν 为清水的动力粘滞系数。

当悬沙浓度较高 ($C_{free} < C < C_{floc} \approx 10 \text{ g/L}$) 时, 处于絮凝沉降阶段, 沉速随悬沙浓度增高而增大, 这时的沉速可按如下根据试验建立的经验公式计算:

$$W_s = K_1 C^{N_1} \quad (8)$$

式中 K_1 为经验常数, N_1 在 1~2 之间, 平均可取为 4/3。

当悬沙浓度很高时 ($C > C_{floc}$), 处于阻碍沉降阶段, 这时沉速随泥沙浓度增大而变小。

$$W_s = K_1 C_{floc}^{N_1} [1 - K_2 (C - C_{floc})]^{N_2} \quad (9)$$

K_2 、 N_2 均为经验参数, N_2 取值一般在 4~5 之间。

1.2. 数值求解方法

采用有限体积法离散求解方程 (1), 方程的数值离散方法与水流方程的离散方法 (见文献[4]) 很相似, 即采用 Roe 的迎风格式结合 MUSCL 方法离散方程中的对流项, 对扩散项则采用中心格式。这样的格式可以保证在浓度具有大梯度时不发生数值振荡而又具有比迎风格式较高的数值精度。

2. 抛泥运动的数值模拟方法

根据抛泥的现场调查发现, 疏浚土进入水体后, 大部分泥沙将以比单颗粒泥沙沉速大得多的速率沉入水底, 这一过程的主要原因是由于高浓度水沙混合体形成异重流。这一过程的泥沙运动机理比较复杂, 因此工程上常常忽略抛泥初期泥沙运动的细节, 而采用远区模型模拟抛泥产生的悬浮泥沙随水流的对流扩散。在这些模型中^{[5][6]}, 是将抛泥带入水体中的泥沙视作泥沙运动方程里的附加源项, 通过将一次抛泥总量平均或加权分配到抛泥区域的各个计算网格以确定抛泥引起的源项强度。在这类方法中, 往往需要假设疏浚土入水以后, 形成悬移质泥沙以及沉降到底的泥沙比例。而实际上这一比例是受具体的受纳环境的水流条件所影响

的, 是动态地变化着。因此这种方法不尽合理。

为了估计抛泥后在抛泥附近区域内泥沙浓度场的变化及初期沉积量分布, 本文采用如下方法: 将抛泥视作瞬时点源引入平面二维模型计算中, 但是不采用直接在模型中添加源汇项的方法, 而是采用在一定的简化条件下由扩散模型解出抛泥一段时间后在受纳水体中的浓度场及淤积量分布, 再叠加到二维模型的计算中去。这些简化假设包括 (1) 所抛泥沙具有与受纳水体的悬沙相同的粒径, 泥沙颗粒均匀, 且均匀沉降; (2) 抛泥点海底平坦, 对悬沙在水体中的浓度分布不产生影响; (3) 抛泥点局部水域的流场在空间上是均匀的, 而且抛泥初始阶段所经历的时间相对较短, 因此在此时段内潮汐流场的变化可以忽略。根据这些简化, 对于排放量为 M 的瞬时源, 文献[7]中解出的由抛泥引起的浓度场及在海底的沉积率分布分别为:

$$C(x, y, z, t) = \frac{M}{8(\pi t)^{\frac{3}{2}} \sqrt{\varepsilon_x \varepsilon_y \varepsilon_z}} \exp \left[-\frac{(x-x_c)^2}{4\varepsilon_x t} - \frac{(y-y_c)^2}{4\varepsilon_y t} - \frac{(z-\bar{z}+w_s t)^2}{4\varepsilon_z t} \right] \quad (10)$$

$$E(x, y, t) = \frac{M(\bar{z}+w_s t)}{16t^{\frac{3}{2}} \pi^{\frac{1}{2}} \sqrt{\varepsilon_x \varepsilon_y \varepsilon_z}} \exp \left[-\frac{(x-x_c)^2}{4\varepsilon_x t} - \frac{(y-y_c)^2}{4\varepsilon_y t} - \frac{(\bar{z}-w_s t)^2}{4\varepsilon_z t} \right] \quad (11)$$

式中 C 为泥沙浓度, M 为初始抛泥量 (kg), ε_x 、 ε_y 和 ε_z 分别是泥沙在 x 、 y 及 z 方向的紊动扩散系数。 w_s 是泥沙沉速, $\bar{z}$ 则是抛泥点距水底的高度, x_c 、 y_c 是抛泥云团中心点的坐标, 它们可根据受纳水体的流场计算。

由此可以得到 t 时刻抛泥产生的单位面积的沉降量为

$$F(x, y, t) = \int_0^t E(x, y, \tau) d\tau \quad (12)$$

$$\text{水深平均的浓度场为: } \bar{C}(x, y, t) = \frac{1}{H} \int_0^H C(x, y, z, t) dz \quad (13)$$

式中 H 为抛泥点水深, $\bar{C}$ 为水深平均的悬沙浓度。

根据 (12) 和 (13) 式就可以给出抛泥一段时间后泥沙在水体中的浓度分布和在水底的沉积分布的一种近似, 在数值模拟时, 我们由以上两式计算出经抛泥经过一段时间 t^* 后 (这时, 进入水体中的泥沙的运动转变为随流输移阶段) 的泥沙浓度和沉积量分布, 再叠加到背景悬移质泥沙的浓度分布中去, 此后就可以按方程 (1) 来计算疏浚土的输移过程。关于抛泥云团运动各个不同阶段的转变, 目前尚无明确的判别条件, 为简化起见, 本文取

$$t^* = \frac{\bar{z}}{w_s} \quad (14)$$

以上就是采用理想瞬时点源扩散模型得到的抛泥数值模拟模型, 实际上, 在疏浚土进入水体时就已经具有一定的初始体积, 而不是一个点源, 可以近似用实际抛泥体上方的一个虚拟点源所发生的扩散过程来代替, 在模型中我们近似取 $\bar{z}=1.1H$ 作为抛泥初始悬浮高度。

3. 澳门周围水域抛泥的模拟

本文以澳门周围水域为算例。澳门周围水域位于珠江口外, 该水域经洪湾水道与珠江磨刀门相连, 水流运动受径流和潮流的相互影响, 其潮汐属不规则半日潮, 日潮不等显著。受地形和径潮流相互顶托的影响, 来自上游西江的泥沙大量淤积在澳门周围水域内, 因此澳门每年都必须定期对个航道进行疏浚, 由航道中清除出来的淤泥则运到水域东南水域开阔处进行倾倒, 如图 1 所示。

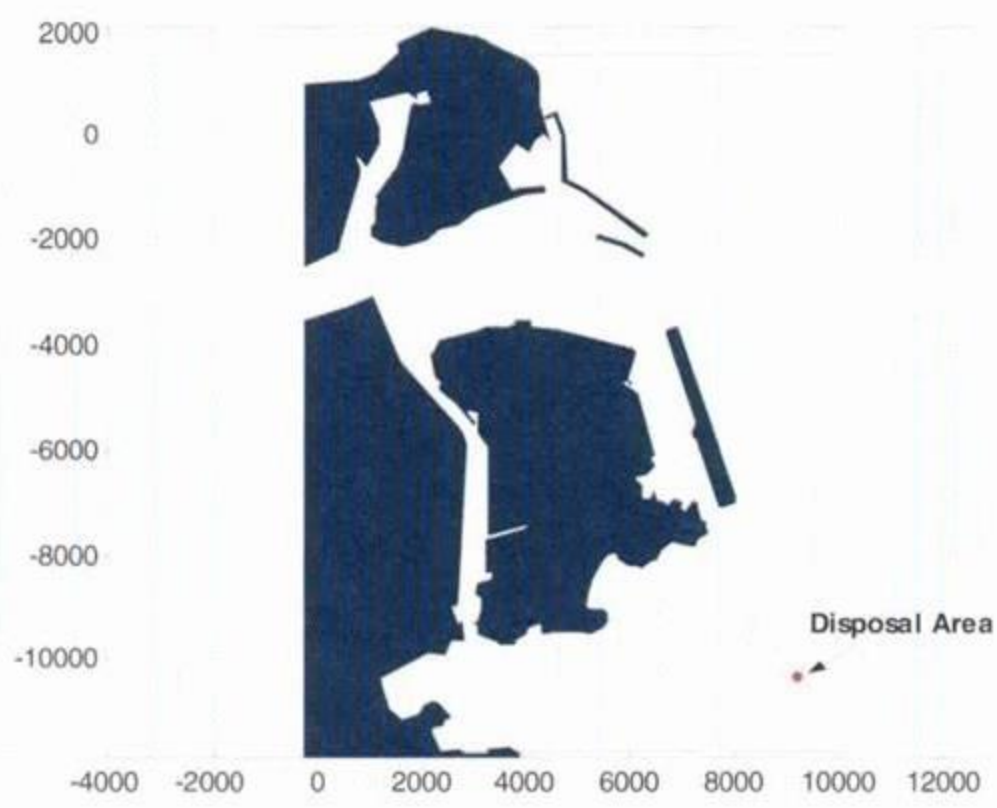
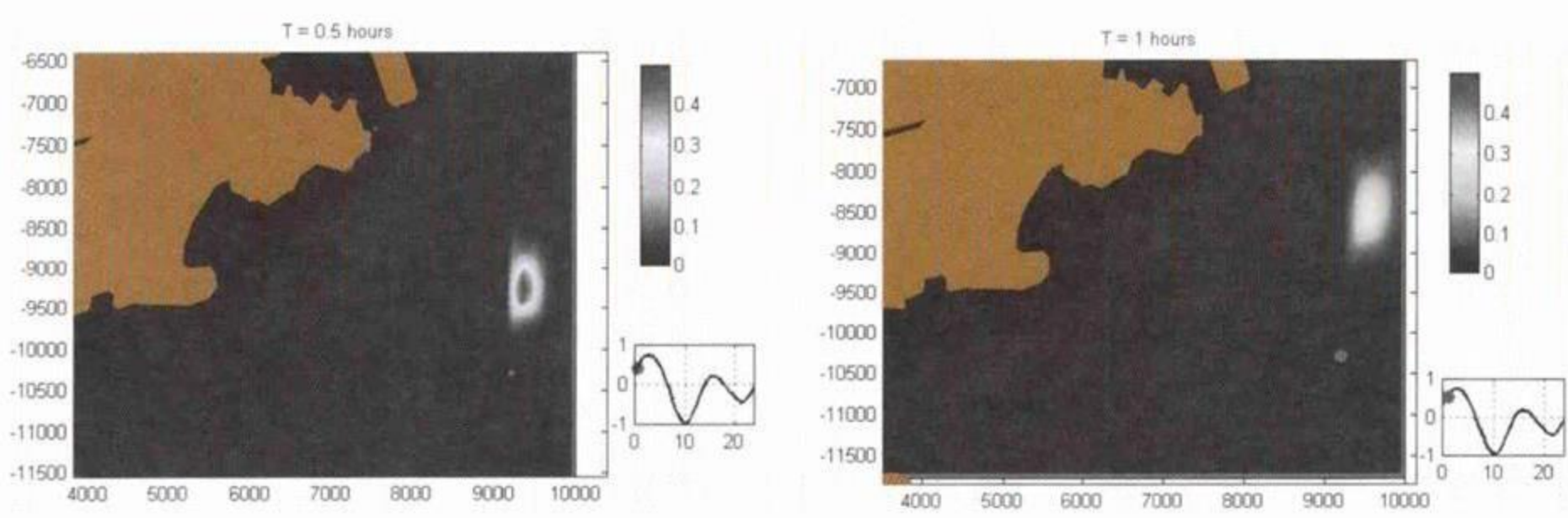


图 1. 澳门周围水域抛泥区位置及计算区域示意图

为了进行抛泥的迁移扩散模拟，首先要正确计算澳门水域的水流及背景悬移质泥沙运动。因此我们首先采用澳门水域 1997 年 9 月 1 日的一组实测水文资料对模型进行了验证计算，该组资料是典型的中水大潮型。验证结果表明，模型计算出的各测站潮位和水深平均流速过程与实测比较吻合，悬移质泥沙过程与实测定性吻合，因此模型可以进行抛泥的迁移扩散模拟计算。



(a) 0.5 小时后 (b) 1 小时后

图 2. 涨潮抛泥扩散分布（浓度单位：kg/m³）

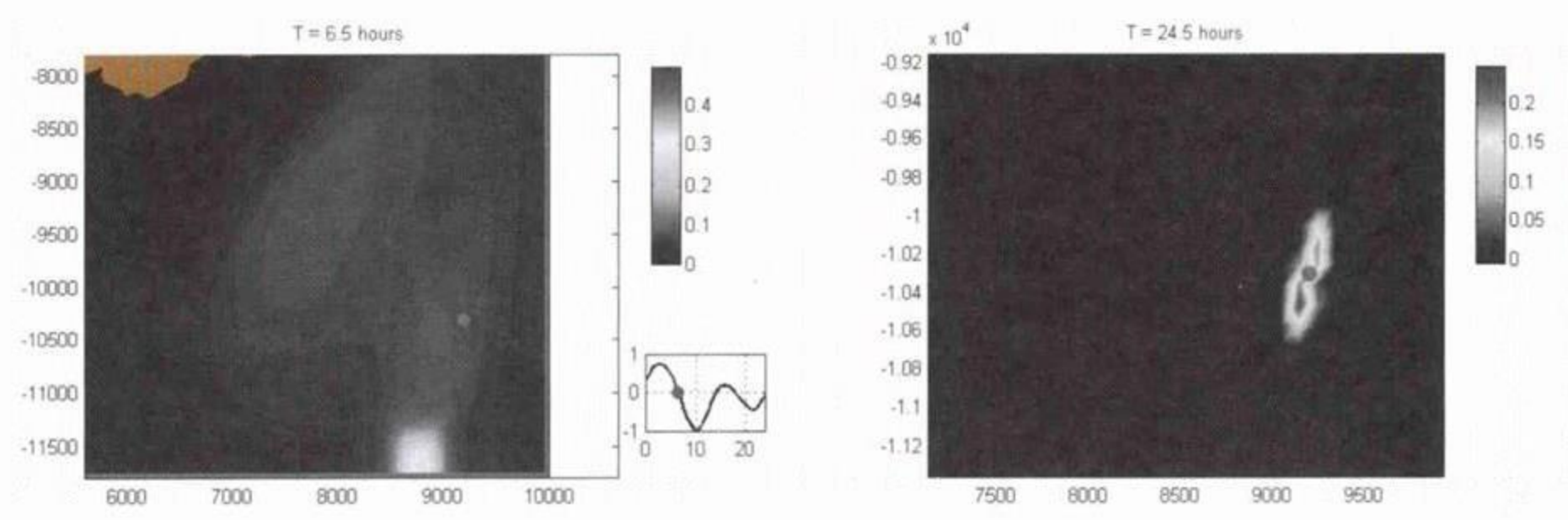


图 3. 落潮抛泥扩散分布（0.5 小时后）

图 4. 抛泥区附近淤积厚度分布（单位：m）

模拟选取的抛泥进行工况如下:每隔 2 小时抛一船泥,单船抛泥体积 3000m^3 ,总共模拟时间为 25 小时。泥沙的紊动扩散系数取 $\varepsilon_x = \varepsilon_y = 1.4\text{m}^2/\text{s}$, $\varepsilon_z = 1.0 \times 10^{-3}\text{m}^2/\text{s}$ 。

图 2 是抛一船泥以后不同时刻抛泥形成的高浓度浑浊带的迁移运动情况(图中粉红色圆点表示抛泥点)。从该高浓度带的输移形态看,它是两个运动的迭加,一是随水流平移,二是相对高浓度中心向四周扩散,但其扩散并非各向同性,而是沿水流方向扩散距离大于其他各向,因此浓度分布呈椭圆型。抛泥后,由于扩散及沿程落淤,高浓度中心的浓度随时间的延续逐渐减小,而扩散范围逐渐增大。选择不同时刻抛泥则扩散情况随水动力条件的不同而有所差异。图 3 给出了在落潮时抛泥的扩散分布情况。由于澳门周围水域的背景悬沙浓度较大,所以抛泥后数小时后抛泥形成的高浓度带最大浓度就降低到跟背景悬沙浓度差不多大,从而也不在有明显的高浓度区域了。

图 4 给出了模拟一天后抛泥区附近的泥沙淤积分布图,由图可见由于抛泥在抛泥区附近产生一个带状的厚淤积区,淤积区的分布走向与潮汐流动的涨潮落潮方向基本一致,淤积量最大的点不是抛泥点而是在抛泥点以南附近,这与抛泥点的水流条件有关。本文计算的淤积形态与以往文献中采用方法^[5]不同,在那种方法中,淤积量最大的点是在抛泥区中心点。由于有水流的流速影响,因此最大淤积量在抛泥中心点不合理。本文的计算结果更合理一些。

4. 结论

采用平面二维泥沙数学模型来研究抛泥形成的悬移质泥沙的长期迁移运动是一个简便有效的手段,本文采用点源扩散模型结合平面二维泥沙数学模型建立了一种模拟抛泥的远区模拟方法。以澳门周围水域为例对抛泥进行了数值模拟。计算结果表明,抛泥在局部水域形成的一个高浓度悬沙带,该高浓度带一方面随涨落潮平移,另一方面由于扩散及淤积,高浓度带的最大浓度逐渐减小,而范围增加。抛泥将引起局部较大的淤积量,但淤积泥沙主要集中在抛泥点附近很小范围内。由于潮汐流动的影响,一般淤积量最大的点不在抛泥点,而是在抛泥点一定距离处。

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澳门水域水柱剖面中有机污染物初步研究

麦碧娴^{1,2}, 杨清书¹, 傅家谟^{1,2}, 盛国英^{1,2}, 彭平安¹

(1. 中国科学院广州地球化学研究所有机地球化学重点实验室, 广州 510640)

(2. 中国科学院广州地球化学研究所广东省环境资源利用与保护重点实验室, 广州 510640)

王志石 邓宇华

澳门大学科技学院, 澳门 3001

摘要: 本研究对采自澳门水域水柱剖面不同深度水体及悬浮颗粒物中的有机污染物进行了分析, 初步结果显示: ①澳门水柱水体中六六六的含量为 8.71-27.02ng/L, 比珠江正干广州河段 (13.8-99.7ng/L) 的低, DDTs 的含量为 8.66-29.76ng/L, 比珠江正干广州河段 (5.0-9.2ng/L) 的高, 16 种优控多环芳烃的含量为 940-6654ng/L, 比珠江正干广州河段 (6558-20031) 低。悬浮颗粒物中六六六的含量为 51.7-3653ng/g, 比文献中珠江口颗粒物中六六六的含量 (1.28-125ng/g) 高, 而 DDTs 的含水量为 180-16236 ng/g, 远远高于珠江口悬浮颗粒物中 DDTs 的含量 (nd-199); ②不同深度水样中有机污染物的组成及分布特征显示, 水体中 DDTs 的浓度及 DDT/(DDE+DDD) 比值从表层往底部减小, 上层样品 (0.5m 和 1.2m) 中 DDT/(DDE+DDD)>1, 可能说明目前仍有新鲜使用的 DDT 农药进入该区水体中; ③悬浮颗粒物中污染物的浓度从表层往底部逐步减小, 污染物在颗粒相和可溶相中的分配系数 (LogKp) 也从上往下具递减趋势, 说明颗粒物主要是以沉降作用和水平迁移运动为主, 底层样品中多环芳烃的 LogKp 值的异常升高, 反映了富含多环芳烃的细颗粒物 (如碳质颗粒) 的底泥再悬浮或是底部海水入浸作用的影响。

关键词: 水柱, 有机氯农药, 多环芳烃

持久性有机污染物 (POPs) 已成为全球环境污染的重要问题, 有关全球环境中 POPs 的含量分布研究表明^[1, 2, 3], 某些 POPs (如 DDTs 和 HCHs 等) 的排放及污染源在最近 20 年已从北半球的工业化国家向南半球的热带—亚热带地区 (如中国和印度) 转移, 在东南亚地区的陆地和海洋环境中 DDTs 的含量最高。由于热带地区的高温和多雨气候条件, 有助于 POPs 向大气快速扩散, 受“全球蒸发分馏模型”的启发^[4], 科学家目前最关心的是“来自热带南亚地区的 POPs 对全球环境中 POPs 的贡献及其在全球 POPs 再循环中的作用”^[3]。

珠江三角洲地处亚热带, 属丰水地区, 境内河网纵横, 雨量丰沛, 各种来源的污染物质将通过地表径流和大气干湿沉降等方式进入珠江河系, 而最终排入珠江口及南海海域, 研究已表明, 位于珠江口西部的澳门周围水域是各种污染物汇集和沉积的场所, 澳门内港沉积物中毒害有机污染物 (DDTs 和 PCBs) 的含量已远远高于沉积物的风险评价标准^[5, 6], 为了进一步探讨该区水体环境中有机污染物的污染状况及底泥沉积物与上层水之间的相与作用, 本研究于 2001 年 4 月在澳门内港采集水柱剖面一条, 对不同深度水体样品中有机氯农药和多环芳烃 (PAHs) 的含量、组成和分布进行了研究, 并根据污染物在水体中颗粒物与可溶相之间的分配, 对污染物在水体中的迁移进行了初步讨论。

1. 样品采集与分析

1.1. 采样

水柱剖面位于澳门水域，采样时间为 2001 年 4 月（枯水期），从表层往底部共设 6 个深度采集，地理位置于 N22°11'41.5"、E113°32'03.5"，样品情况如表 1 所示，表层（0.5m）至 5.5m 深水样采用一般水泵采集，为了保证不扰动表层底泥，底部（沉积物与水界面）水样采用蠕动泵采集，每个水样为 50l，装于干净棕色玻璃瓶内，运往实验室，水样过 GF/F 玻璃滤膜收集颗粒物，XAD-4 和 XAD-2（1:1）树脂富集水相中的有机质，分别对颗粒物和水中相的多环芳烃和有机氯农药进行了定量分析，

1.2. 多环芳烃和有机氯农药的分析

滤膜颗粒物冷冻干燥后称重，加入一定量的蔡-d₈、二氢苊-d₁₀、菲-d₁₀，屈-d₁₂、北-d₁₂ 和 4，4 二氯联苯作回收率指示物，二氯甲烷索氏提取 48 小时。富集了有机质的 XAD 树脂，注入一定量的回收率指示物后，先用甲醇洗脱，然后用甲醇:二氯甲烷（1:1）超声或索氏萃取未洗脱的残余有机质，合并洗脱液和萃取液，加入一定量的干净水，用二氯甲烷液一液萃取，收集二氯甲烷萃取液，对颗粒物和树脂的萃取液分别浓缩并溶剂替换为正己烷后，过硅胶/氧化铝（2:1）层析柱，用 15ml 正己烷和 70ml 二氯甲烷/正己烷（3:7）分别淋洗出烷烃和芳烃组分，洗脱液浓缩定容后（0.4ml），再加入一定量的六甲基苯和 2,4,5,6 -四氯间二甲苯及十氯联苯作内标，分别进行 GC-MSD 和 GC-ECD 分析。

表 1 澳门水域水柱样品采样情况

样号	深度(m)	盐度	浊度	PH	悬浮物含量（mg/L）
MW-1	0.5	3.0	11.7	6.7	1.7
MW-2	1.2	3.0	12.0	6.7	17.9
MW-3	3.5	10.0	21.6	6.7	42.5
MW-4	4.8	14.0	41.9	6.7	54.0
MW-5	5.5	15.0	49.2	7.0	58.5
MW-6	6(底部)	15.0	62.1	7.0	73.4

1.3. 化合物的定量

化合物的定量采用 5 点校正曲线和内标法进行。有关标准物质、PAHs 的气相色谱-质谱联用仪（GC-MSD）分析和有机氯农药及多氯联苯的气相色谱-电子捕获检测器（GC-ECD）分析和方法检测限等，请参阅文献^[6]。

1.4. 质量控制和质量保证（QA/QC）

整个分析过程由以下 USEPA 的 QA/QC 控制样监控：方法空白、加标空白、基质加标、基质加标平行样、样品平行样，并用回收率指示物监测样品的制备和基质的影响。本研究颗粒物样品中指示物的回收率(mean±stdev)分别为：蔡-d₈：47.6±8.2%，二氢苊-d₁₀：75.6±23.5%，菲-d₁₀：89.1±6.2%，屈-d₁₂：95.2±3.3%，北-d₁₂：98.0±16.7%，4，4-二氯联苯：95.2±9.6%，溶解相样品中回收率指示物的回收率(mean±stdev)分别为：蔡-d₈：36.6±5.2%，二氢苊-d₁₀：66.7±20.7%，菲-d₁₀：82.1±7.2%，屈-d₁₂：92.2±6.3%，北-d₁₂：95.2±11.4%，4，4-二氯联苯：88.2±8.8%，化合物的定量结果经回收率校正。详细的分析方法及质量控制/质量保证参看文献^[6]。

2. 结果与讨论

2.1. 有机氯农药

2.1.1. 水柱剖面中有机氯农药含量及污染程度初步分析

表 2 列出了水柱样品中 21 种有机氯农药的定量结果，结果显示：所有目标化合物在水体中均有检出，水体中六六六的含量为 8.71-27.01ng/l，比珠江正干广州河段（13.8-99.7ng/l）的低，DDTs 的含量为 8.66-29.76ng/l，比珠江正干广州河段（5.0-9.2ng/l）的高，其它有机氯农药包括：艾氏剂、狄氏剂、异狄氏剂、硫丹 I、异狄氏剂醛、异狄氏剂酮、硫丹硫酸盐、硫丹 II、七氯、七氯环氧化物和甲氧滴滴涕，其含量范围为：7.7-25.19ng/l，比珠江正干广州河段（17.46-61.56ng/l）的低。表 1 同时列出了我国《地表水环境质量标准》（GHZB1-1999）和美国 EPA 地表水水质标准（EPA822-Z-99-001）中对有机氯农药的限值。从表中可以看到，水体样品中 p,p'-DDT、p,p'-DDE、p,p'-DDD 和艾氏剂的浓度大大超过美国 EPA 地表水质标准限值，其中 DDT 和 DDD 化合物的含量（2.15—13.11ng/l）比水质标准高一个数量级（0.59—0.83ng/l）。其它超过标准限值的化合物有：α-六六六、七氯、七氯环氧化物、狄氏剂、异狄氏剂。水体悬浮颗粒物中六六六的含量为 51.7-3653ng/g，比文献^[7]中珠江口颗粒物中六六六的含量（1.28-125ng/g）高，而 DDTs 的含量为 180-16236 ng/g，远远高于珠江口悬浮颗粒物中 DDTs 的含量（nd-199）。尽管我国在 1982 年已停止使用六六六和 DDT 等有机氯农药，但其在环境中的残留及难降解性，在澳门内港水体中的高浓度的检出，应引起重视。

表 2 澳门水域水柱样品中有机氯农药的定量结果（ng/l）

化合物	样 品 号						地表水质量标准值 (GHZB 1-1999)	EPA 推荐标准 (EPA822-Z-99-001)
	MW-1	MW-2	MW-3	MW-4	MW-5	MW-6		
α-六六六	4.48	6.80	4.24	2.85	1.98	6.26		3.9
β-六六六	3.08	4.85	3.35	1.80	2.11	4.06		14
γ-六六六(林丹)	5.33	7.89	4.33	3.26	2.22	7.66	19	19
σ-六六六	5.37	7.48	4.66	3.23	2.39	8.69		
六六六 总量	18.26	27.02	16.57	11.14	8.71	26.67	5000	
p,p'-滴滴涕	13.11	10.12	3.48	5.40	2.15	3.10		0.59
p,p'-滴滴伊	1.49	1.04	1.09	0.85	0.62	1.47		0.59
p,p'-滴滴滴	10.13	6.72	6.55	8.93	4.41	7.88		0.83
o,p'-滴滴涕	3.50	2.88	2.08	2.03	0.90	2.14		
o,p'-滴滴伊	0.25	0.16	0.10	0.10	0.14	0.15		
o,p'-滴滴滴	1.28	0.93	0.81	0.78	0.44	0.94		
滴滴涕总量	29.76	21.85	14.11	18.09	8.66	15.67	1000	
七氯	0.93	0.44	1.52	0.50	0.56	1.18		0.21
艾氏剂	5.14	1.73	3.34	4.24	2.26	3.04		0.13
七氯环氧化物	0.39	0.11	0.21	0.43	0.53	4.57		0.1
硫丹 I	3.26	3.70	4.41	1.34	2.26	4.16		110000
狄氏剂	0.41	0.16	0.58	0.34	0.39	1.48		1.4
异狄氏剂	2.07	0.31	0.69	4.80	0.51	2.68		2.3
硫丹 II	1.12	1.29	2.28	0.59	0.49	3.00		110000

表 2（续） 澳门水域水柱样品中有机氯农药的定量结果（ng/l）

化合物	样 品 号						地表水质量标准值 (GHZB 1-1999)	EPA 推荐标准 (EPA822-Z-99-001)
	MW-1	MW-2	MW-3	MW-4	MW-5	MW-6		
异狄氏剂醛	2.12	0.59	0.35	1.46	0.02	2.30		760
硫丹硫酸盐	0.12	0.02	0.22	0.57	0.51	0.77		110000
异狄氏剂酮	0.83	0.06	0.65	0.72	0.13	0.54		
甲氧滴滴涕	0.15	0.95	0.10	0.08	0.05	1.47		30
其它含氯农药总量	16.54	9.36	14.35	15.06	7.70	25.19		

表 3 澳门水域水柱样品悬浮颗粒物中有机污染物的浓度（ng/g）

样号	HCHs	DDTs	其它含氯农药	16PAHs
MW-1	3653.0	16235.3	5806.5	88429.6
MW-2	448.3	1125.8	260.3	5165.6
MW-3	137.2	269.5	193.5	6024.2
MW-4	93.9	325.6	208.1	3650.4
MW-5	51.7	139.0	87.4	2564.9
MW-6	59.4	180.0	238.6	3452.6

2. 1. 2. 水柱剖面中 DDT 及其降解产物的垂向变化

DDE 和 DDD 是 DDT 的降解产物,DDT 在厌氧条件下通过土壤微生物的作用转化为 DDD, 在好氧条件下则转化为 DDE^[8], 因此使用 DDT/（DDD+DDE）和 DDD/DDE 比值, 可以指示 DDT 的降解程度、输入情况及降解过程的氧化还原条件。图 1 显示了澳门内港水柱样品中 DDT/（DDE+DDD）和 DDD/DDE 比值的变化, 从图中可以看到, 水柱中 DDT/（DDE+DDD）比值从表层往底部减小, 上层两个样品（0.5m 和 1.2m）中 DDT/（DDE+DDD）>1, 可能说明目前仍有新鲜使用的 DDT 农药进入该区水体中, DDD/DDE 比值较大, 说明主要为还原环境, 这与该区的高浊度水体有关

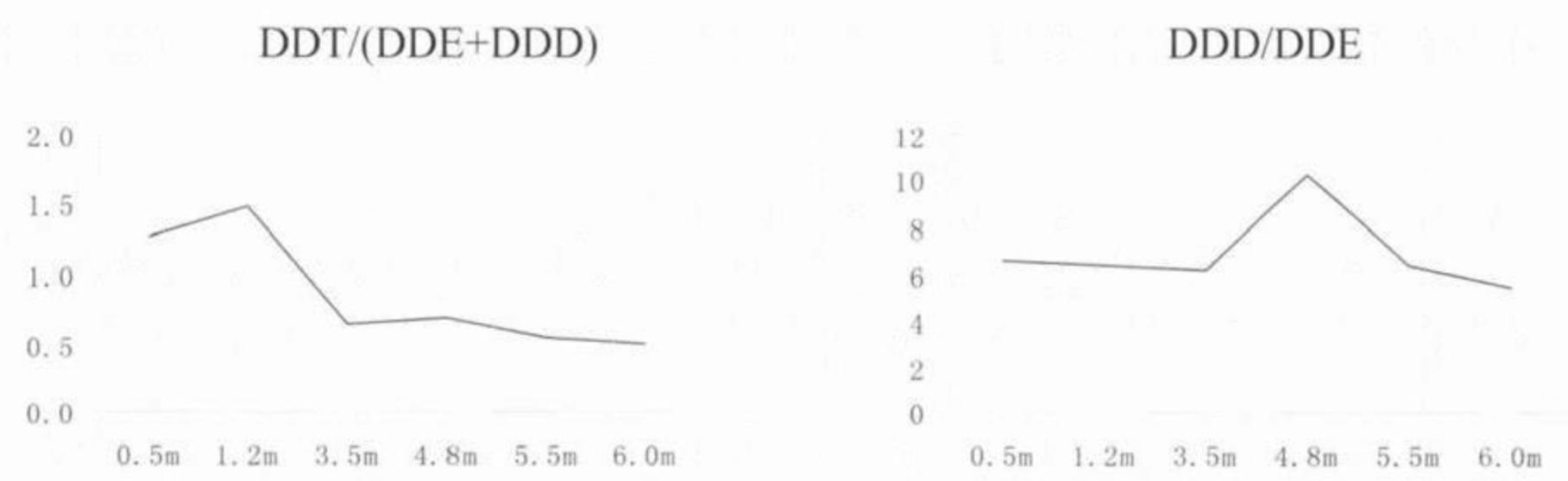


图 1 水柱剖面中 DDT/（DDE+DDD）和 DDD/DDE 比值的变化

2. 1. 3. 有机氯农药在水体溶解相和颗粒相中的分配

图 2 显示了各种有机氯农药在不同深度样品颗粒相和可溶相中的分布情况, 从图可以看到, 位于水柱 1.2m 和底部的水体中六六六的浓度最高, 而且可溶相中的六六六的浓度比颗粒物中的高, 水体中 54.5—83.7% 的六六六以可溶相状态出现, 表层水

(0.5m) 中六六六的浓度比下部水样 (1.2m) 的低, 这可能是由于六六六的蒸气压较高, 表层水中的六六六向大气挥发的结果, 底部界面水中六六六的浓度也较高, 而且主要存在于可溶相 (83.7%) 中, 这可能与底泥中六六六的溶解再释放作用有关。水体中 DDTs 的浓度从表层往底部呈现减少的趋势, 而且 84.3—97.2% 的 DDTs 赋存于悬浮颗粒物中, 这一方面与 DDTs 和溶解度低有关, 另一方面表明该区水域目前水体中的 DDTs 主要来自周围及上游农业地区土壤的风化侵蚀, 上层水体高的 DDT/(DDE+DDD) 比值, 说明某些地区可能仍然少量使用 DDT 等有机氯农药。

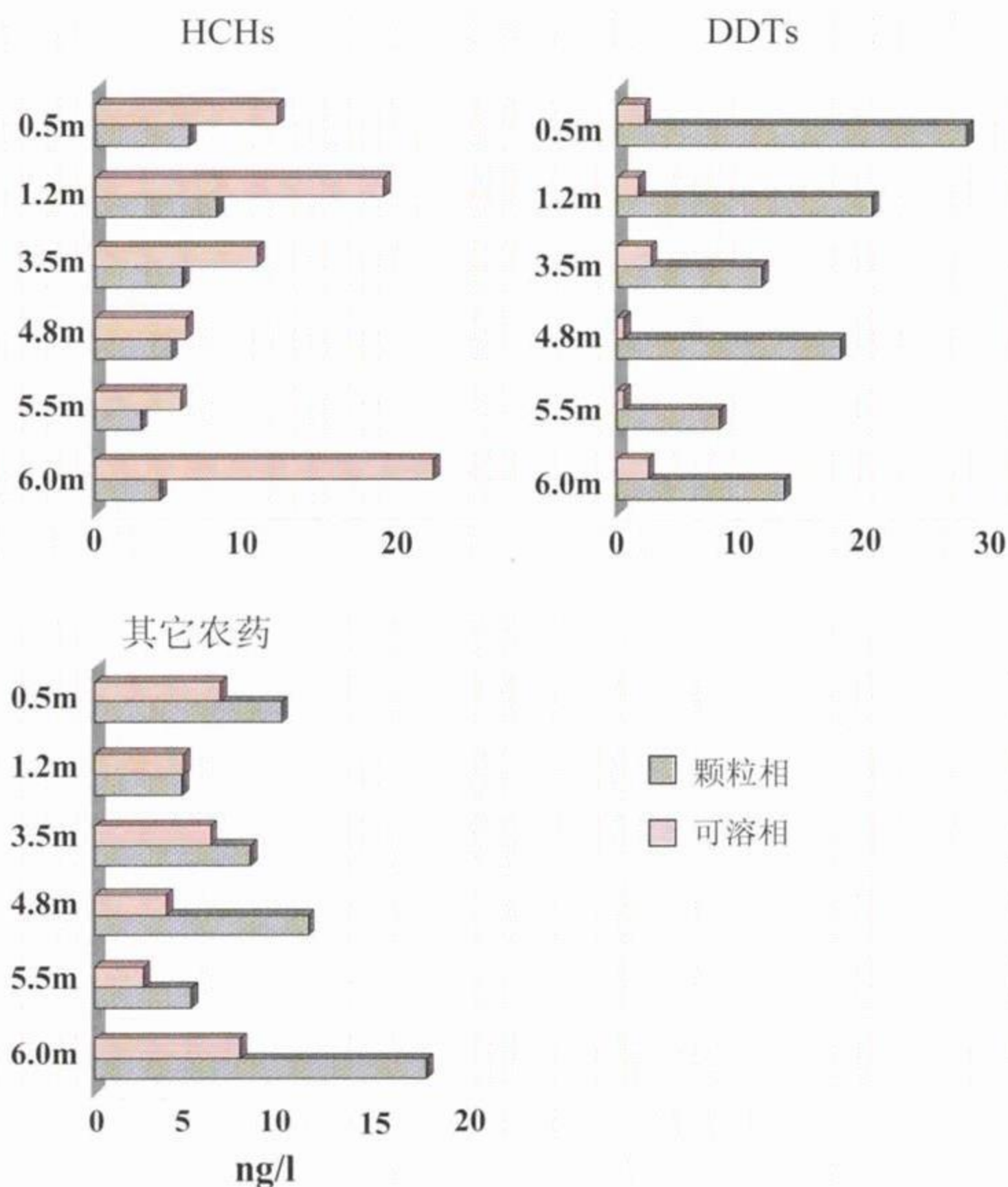


图 2 澳门内港水柱剖面中有机氯农药的纵向分布

从悬浮颗粒物中有机氯农药的浓度梯度 (图 3) 看, 从表层往底部有机氯农药的浓度减小, 说明颗粒物主要以沉降作用和水平迁移运动为主, 没有表现出明显的泥质再悬浮作用特征。从有机氯农药在颗粒相和可溶相中的分配系数 (LogKp) 来看, 有机氯农药的 LogKp 值从上往下具递减趋势 (图 4), 说明水体中颗粒物对有机氯农药的富集能力从上往下减小, 这是从上往下颗粒物粒度变粗、有机质含量减小的结果。

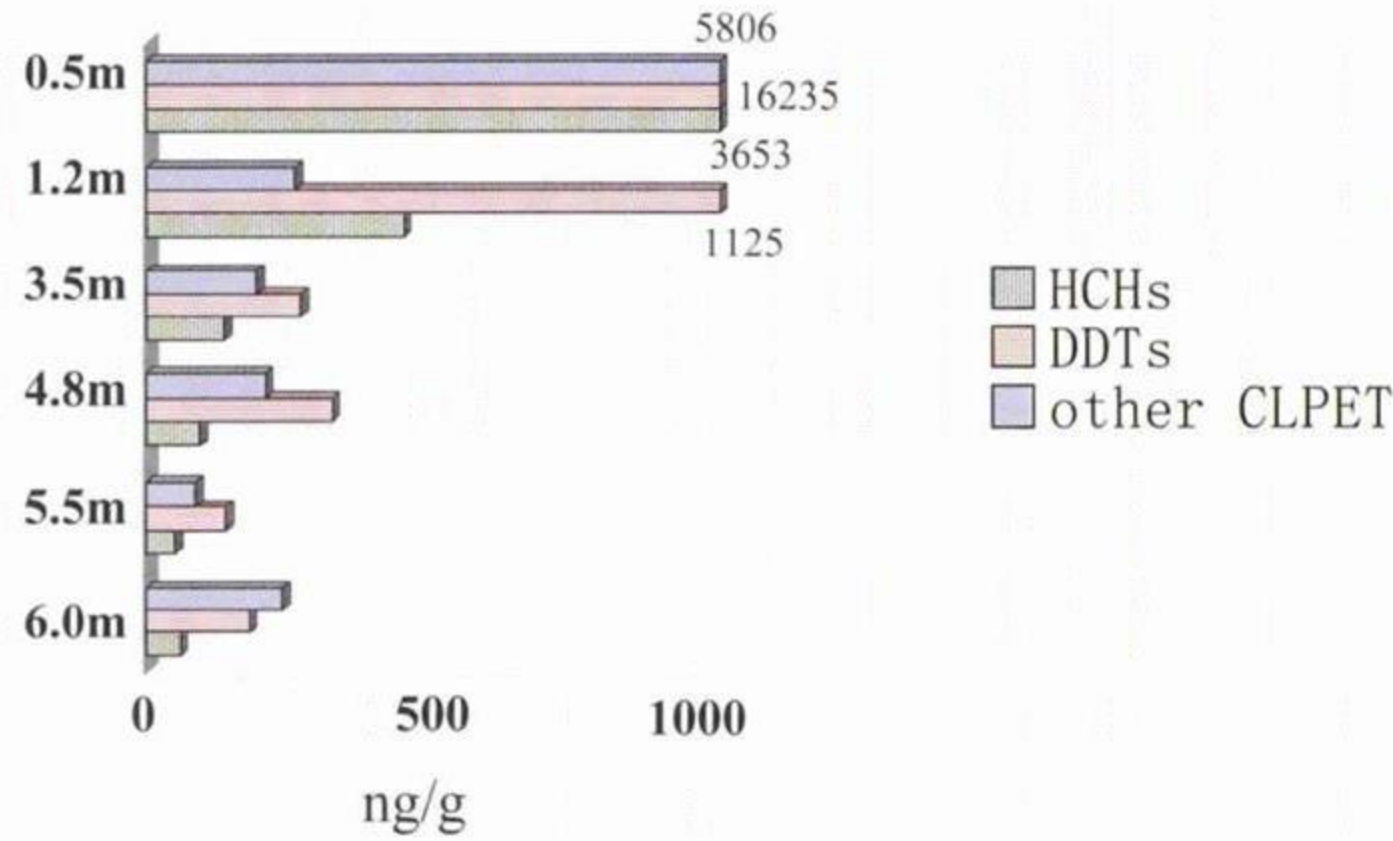


图 3 澳门内港水柱颗粒物中有机氯农药的浓度分布

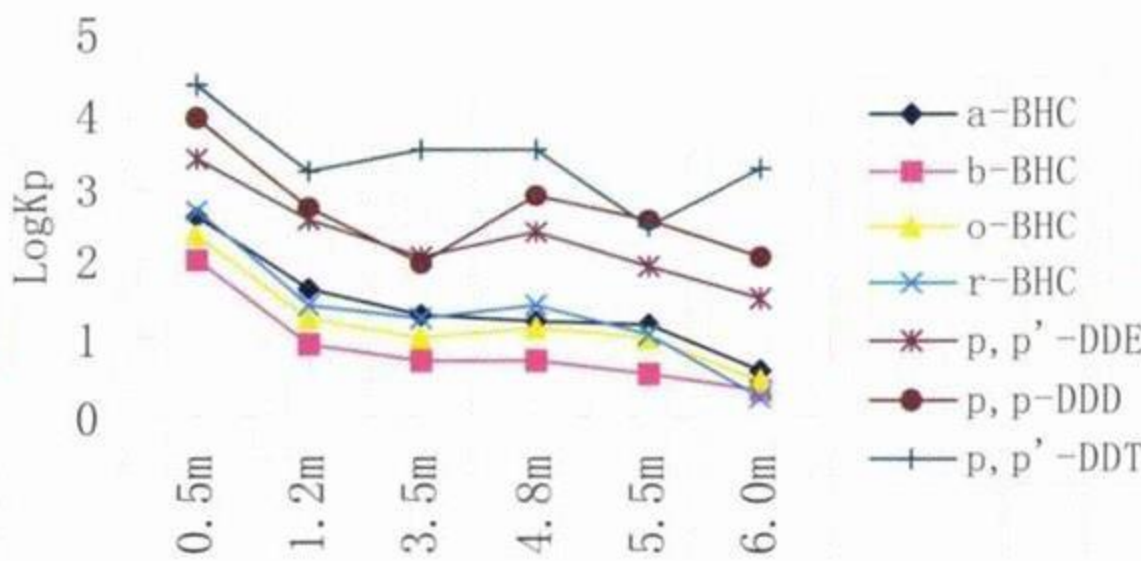


图 4 农药在澳门水柱中颗粒物和水中相的分配系数 (LogKp)

2.2. 多环芳烃

2.2.1. 水柱剖面中多环芳烃含量及污染程度初步分析

表 3 列出了水柱样品中 16 种优控多环芳烃的分析结果, 16 种多环芳烃的含量从高到低依次为: 蔡>>菲>芴>荧蒹、芘>芘烯、屈>蒽、苯并(b)荧蒹>芘、苯并(ghi)芘、苯并(a)蒽>二苯并(a,h)蒽、茚并(1,2,3)芘、苯并(k)荧蒹、苯并(a)芘。16 种优控多环芳烃的总量为 940-6654ng/L, 比珠江正干广州河段 (6558-20031) 低。水体样品中屈和苯并(b)荧蒹的含量超过美国 EPA 地表水质标准限值, 但强致癌化合物苯并(a)芘的浓度低于标准限值。

表 3 澳门水域水柱样品中多环芳烃的含量 (ng/l)

化合物	样 品 号						地表水标准值 (GHZB 1-1999)	EPA 推荐标准 (EPA822-Z-99-0 01)
	MW-1	MW-2	MW-3	MW-4	MW-5	MW-6		
蔡	5992.62	4255.83	4703.7 2	6447.50	1529.7 9	767.26		
芘	4.98	1.97	2.48	3.35	1.21	1.22		
芘烯	5.06	3.88	11.39	10.97	6.98	14.33		1200000
芴	41.83	19.27	47.16	40.58	21.78	36.79		1300000
菲	101.82	33.51	93.37	79.90	44.01	68.73		

蒽	6.38	2.84	8.00	7.89	4.19	5.64		9600000
荧蒽	10.23	7.18	19.66	14.97	13.40	14.02		300000
芘	14.23	6.63	16.42	15.07	12.06	10.55		960000
苯并(a)蒽	1.98	1.23	3.21	3.55	1.94	1.57		4.4
屈	9.63	6.16	9.94	10.20	9.01	7.77		4.4
苯并(b)荧蒽	4.52	4.03	7.70	8.24	6.99	4.32		4.4
苯并(k)荧蒽	1.15	1.00	1.83	1.99	2.37	1.51		4.4
苯并(a)芘	1.17	0.94	1.52	1.99	1.65	1.25	2.8	4.4
茚并(1,2,3)芘	1.36	1.09	2.37	2.60	2.25	1.60		4.4
二苯并(a,h)蒽	1.30	0.91	2.37	2.86	2.45	1.35		4.4
苯并(ghi)芘	2.34	1.65	2.54	2.46	2.56	2.15		
总量	6200.6	4348.1	4933.7	6654.1	1662.6	940.1		

2.2.2. PAHs 组成及来源初探

图 5 为低分子量母体多环芳烃（分子量为 178-202）/高分子量母体多环芳烃（分子量为 226-300）（LMW-P-PAHs/HMW-P-PAHs）比值与烷基化多环芳烃/母体多环芳烃（Alkyl-PAHs/Parent-PAHs）比值图，从样品在该图的位置，可以显示样品中多环芳烃的组成及有关来源的信息，受油类排放多环芳烃污染的样品，具有高的烷基化多环芳烃/母体多环芳烃比值，而受高温燃烧来源多芳烃污染的样品具有低的烷基化多环芳烃/母体多环芳烃比值和低的低分子量母体多环芳烃/高分子量母体多环芳烃比值，从图 5 水柱样品显示的情况可以看到，水体样品中具有很低的烷基化多环芳烃/母体多环芳烃比值（0.10—0.65），与气溶胶样品中的比值（0.01—0.52）接近，而低分子量母体多环芳烃/高分子量母体多环芳烃比值相对较高（1.23—2.91），比该区其它环境样品（沉积物、大气气溶胶和降尘）的高。水体样品中多环芳烃的这些组成特征表明，水体中的多环芳烃主要为燃烧来源的产物。从图 5 水体颗粒物样品所处的位置可以看到，水体颗粒物的组成与澳门周围水域沉积物中多环芳烃的组成相似。

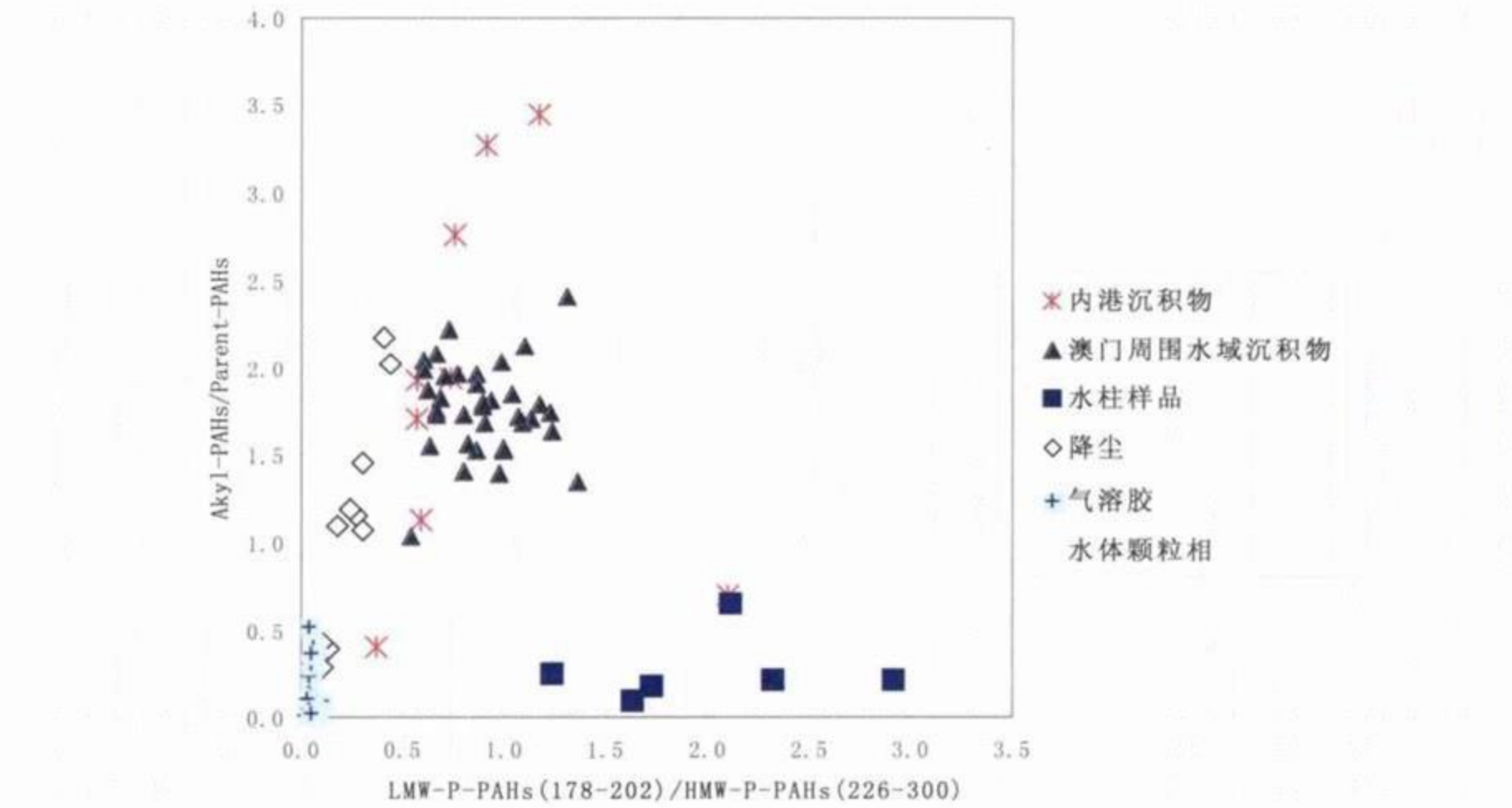


图 5 多环芳烃中低分子量母体多环芳烃/高分子量母体多环芳烃比值与烷基化多环芳烃/母体多环芳烃比值图

2.2.3. PAHs 在水体溶解相与颗粒相中的分配

图 6 显示了不同环数多环芳烃在水柱样品可溶相和颗粒相中的分布情况,不同环数多环芳烃在水柱剖面中的分布特征不同,水体中 2 环多环芳烃的含量上部高,下部低,而且 86.9—99.5% 的 2 环多环芳烃呈溶解态出现,2 环多环芳烃含量的纵向分布特征表明水体中的 2 环多环芳烃可能是通过水—气交换方式进入水体的。3 环多环芳烃含量分布变较大,在表层 (0.5m)、0.6m、0.8m 和界面水体中较高,而在 0.2m 和 5.5m 水体中较低,在各采样层位颗粒相与溶解相的分布也不同,在沉积物与水体的界面水中仅有 12.9% 的 3 环多环芳烃以可溶态存在,而在表层水中,则有 63.5% 的 3 环多环芳烃存在于可溶态中。4 和 5-6 环多环芳烃在上层 (0.5m 和 1.2m) 水中的含量比下层水低,且主要存在于颗粒物中,其中 78.0%-95.9% 的 4 环多环芳烃和 89.7-99.7% 的 5-6 环多环芳烃赋存在颗粒物中,悬浮颗粒物中 4-6 环多环芳烃的浓度梯度变化 (图 7) 与有机氯农药的变化超势相同,从上往下逐步减小,进一步说明颗粒物主要以沉降作用和水平迁移运力为主。而多环芳烃污染物的 LogKp 沿深度变化较有机氯农药的变化复杂 (图 8),在上部的 4 个采样层位 (表层—4.8m) 其 LogKp 值呈递减,这反映颗粒物的正常沉降过程,而在底层 (5.5m) 和底部的水样中,LogKp 值的变化发生了异常,底层 (5.5m) 水中 LogKp 值比底部水的高,说明底层水中相对富集了一些易于吸附多环芳烃的细颗粒物 (如碳质颗粒),这可能是底泥再悬浮或者是底部入浸海水所携带细颗粒物补充的结果。

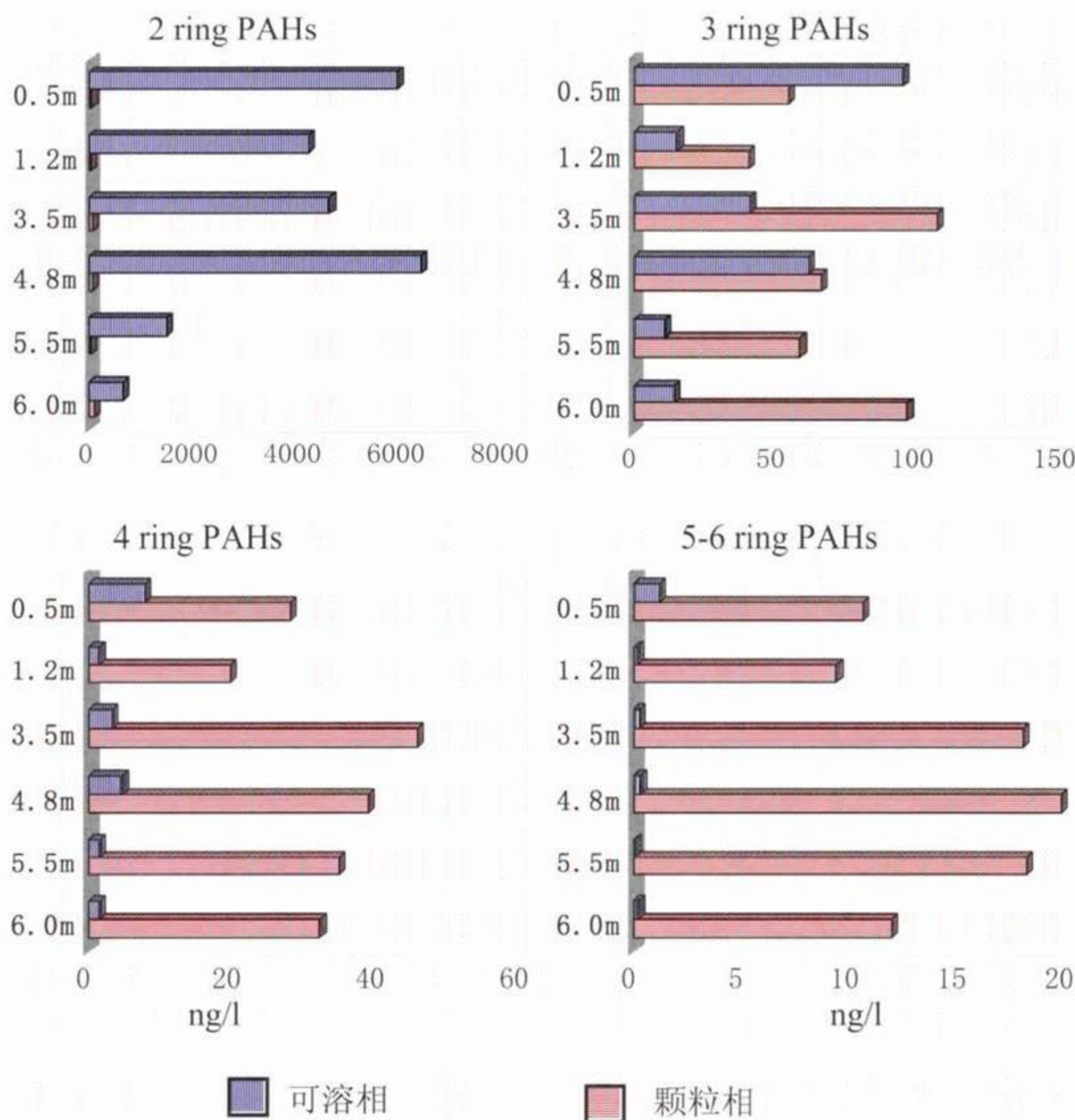


图 6 澳门内港水柱剖面中多环芳烃的纵向分布

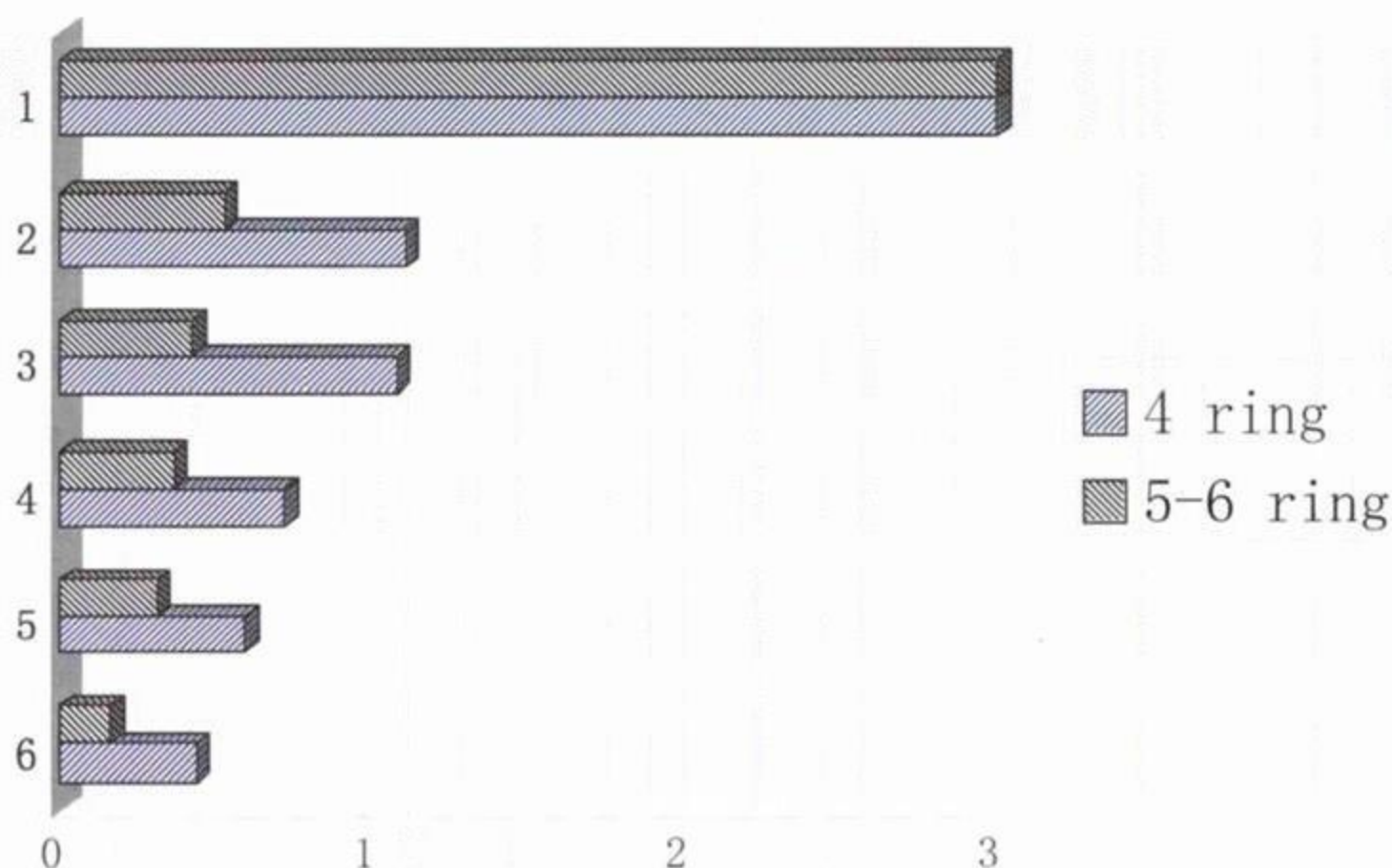


图 7 澳门内港水柱颗粒物中多环芳烃的浓度分布

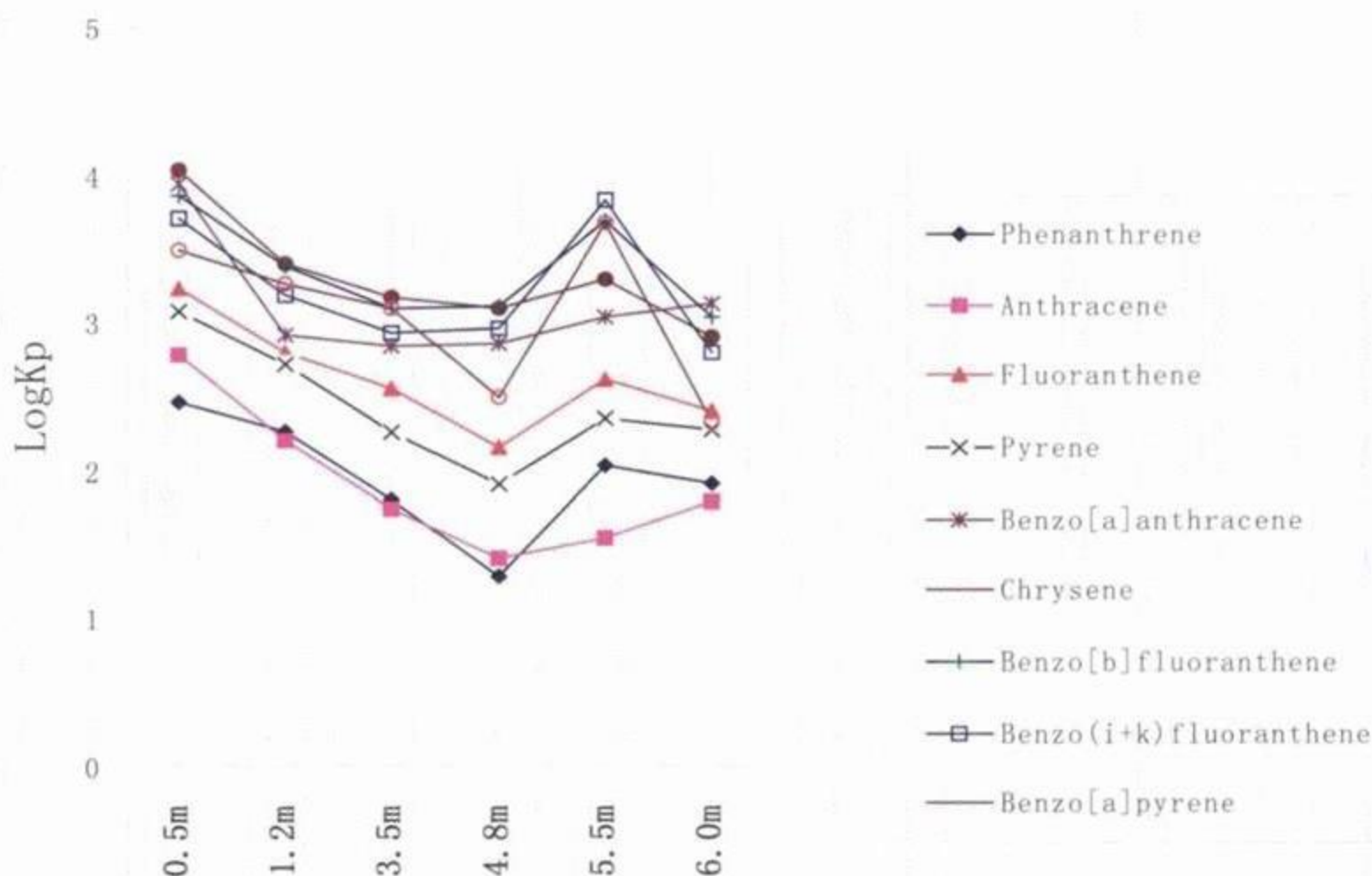


图 8 多环芳烃在澳门水柱中颗粒物和水中相中的分配系数 (LogKp)

3. 初步结论

(1) 澳门内港水体中有机氯农药 DDTs 的含量较高, 部分有机氯农药的浓度大大超过 EPA 的地表水质标准值, 而且目前可能仍有少量新使用的 DDT 农药输入;

(2) 水体中多环芳烃的污染程度比珠江广州河段低, 但仍有个别多环芳烃化合物的含量超过 EPA 的地表水质标准值, 水体中多环芳烃主要为不完全燃烧的产物。

(3) 水体中颗粒物主要以沉降和水平迁移运动为主, 底泥的再悬浮作用只对底层 (从界面向上 0.5m±) 水体产生一定的影响。

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Preliminary study of organic pollutants in the water column from Macao coastal waters

MAI Bixian^{1,2}, Yang Qisu¹, Fu Jianmon^{1,2}, SHENG Guoying^{1,2}, and PENG Pingan¹

(1. State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou, 510640, China

2. Guangdong Key Laboratory of Environmental Protection and Resources Utilization, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou, 510640, China)

Zhishi Wngg, U Wa Tang

Faculty of Science and Technology, University of Macao, Macao

Abstract: The organochlorine pesticides and polycyclic aromatic hydrocarbons (PAHs) in water column from Macao coastal waters were analyzed quantitatively, based on USEPA 8000 series methods and under quality assurance and quality control (QA/QC). The results are as follows: (1) The concentrations of HCHs and 16 PAHs in Macao coastal waters is range from 8.71 to 27.02 ng/l and 940-665ng/l, respectively, lower than that in Pearl River waters. Howere, the DDTs levels is higher that from Pearl River, ranging from 8.66-29.76ng/l. The HCHs and DDTs levels in suspended particles Macao costal waters is much higher than that from the Pearl River estuary. (2)The concentrations of DDTs and the ratios of DDT/(DDE+DDD) decrease with increasing depth in the water colum. The DDT/(DDE+DDD) in the suface water samples is higher than 1. These charateristics suggest that there are still fresh inputs of DDT into the Macao coastal waters. (3) The concentrations of organic pollutants in suspended particles and the partition coefficients (LogKp) between particle phase and dissolved phase are decreasing with increasing depth, indicating predominat deposition process and horizontal transportation for particles in water column. The relative high LogKp values are found for PAHs in deeper samples, suggesting the contribution of particle-bound PAHs derived from resuspended sediments and suspend particals in sea water.

Keywords: water column; organochlorine pesticides, polycyclic aromatic hydrocarbons(PAHs)

澳门南湾湖的环境问题与对策分析*

杜鹏飞 曾思育 陈吉宁

(清华大学环境科学与工程系, 环境模拟与污染控制国家重点联合实验室, 北京 100084)

王志石, 邓宇华

(澳门大学科技学院, 澳门)

摘要: 南湾湖是澳门本岛唯一的淡水湖泊, 也是重要的景观水体。通过对澳门南湾湖进行实地考察并分析长期监测数据, 可以将南湾湖的环境问题归纳如下: (1) 水质污染; (2) 富营养化; (3) 水草过盛; (4) 垃圾影响水质及景观; (5) 监督与管理薄弱。鉴于此, 应重点从以下方面入手, 解决南湾湖的环境问题: (1) 加强监测和监督; (2) 对污染源进行细致调查, 从源头削减污染物的排放; (3) 针对水草问题开展研究, 寻找适宜的控制对策; (4) 加强制度建设; (5) 开展规划研究, 因地制宜地确定控制目标和管理办法。

关键词: 南湾湖, 富营养化, 水草, 环境管理

1. 南湾湖的功能定位

南湾湖位于澳门本岛南端, 分东西两个湖体, 东湖面积约 0.45Km², 西湖面积约 0.24Km²。该湖泊是澳门地区唯一的淡水湖泊, 地处著名的妈阁山南麓, 有中国银行、普京大酒店、澳门立法委大楼等重要建筑伫立在湖畔, 嘉乐庇总督大桥跨湖而过, 连通了澳门本岛与氹仔岛, 投资 1.12 亿美元、高达 338 米的观光塔在两湖之间拔地而起, 再加上一年一度的端午节龙舟竞赛, 使这里成为澳门地区重要的旅游观光景区。



图 1 景色迷人的 澳门南湾湖

来源: http://desktop.china.com/zh_cn/cool/view/macao/

2. 南湾湖的水环境质量

根据南湾湖的水体功能定位, 采用国家《地表水环境质量标准 (GHZB1-1999)》中的 IV 类水体标准进行评价。表 1、2 所示分别为 2000 年和 2001 年的监测结果。

表 1 2000 年澳门南湾湖水体水质指标监测平均值

Table 1 Mean values of water quality monitoring data of Nam Van Lake

指标	pH	COD _{Cr}	BOD ₅	亚硝酸盐	酚类	硝酸盐	溶解氧
I湖平均	8.25	88.74	19.30	0.03	6.17	1.05	6.46
II湖平均	8.30	35.22	7.14	0.04	2.50	1.32	6.53

* 本研究得到澳门市政厅及澳门大学的资助。

IV类水标准	6.5-8.5	30	6	1.0	0.01	20	3
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表 2 2001 年澳门南湾湖水体水质指标监测平均值

Table 2 Mean values of water quality monitoring data of Nam Van Lake						
指标	pH	I _{Mn}	总磷	亚硝酸盐	硝酸盐	溶解氧
I湖平均	9.0	4.67	0.0819	0.004	1.32	6.53
II湖平均	8.4	7.36	0.0379	0.004	1.05	6.46
IV类水标准	6.5-8.5	10	0.2	1.0	20	3

评价结果表明：

(1) 目前南湾湖水体已经受到污染，多数指标满足 IV 标准，但个别指标(如 COD 等)甚至劣于 V 类标准，水体质量总体评价结论为劣于 V 类，污染特征为有机型污染，I 湖水水质要略差于 II 湖水水质。

(2) 南湾湖已经面临着富营养化问题，富营养化状况属富营养级别(富营养化评价中的 IV 类标准)，总磷和悬浮物是主要的污染物，总氮可能是南湾湖富营养化的限制因子。



图 2 南湾湖的区位分析

3. 南湾湖的水草问题

2000 年 12 月和 2001 年 5 月，笔者对南湾湖进行过两次现场踏勘，并询问了当地居民及清运水草、垃圾的工人。据估算，在水草生长旺盛期，南湾湖每天清除水草约 70 吨。



图 3 南湾湖 I 湖中捞出的水绵体



图 4 南湾湖 II 湖中漂浮的金鱼藻

一个良好的水生生态系统应包括四个基本组成成分：无机环境、生物生产者、消费者和分解者。藻类在水生生态系统中作为生产者对维持生态平衡起着至关重要的作用，藻类通过吸收水体中的营养物质和水分，在阳光下通过光合作用转化为生物能，并借助于食物链流向动物和微生物。以藻类和其他生物为核心的湖泊生态系统，是湖泊水生生态系统员基本的特征。但是，过度生长的藻类，不仅表征着湖泊的污染，而且也破坏了湖泊水体中自然生态系统中的自我控制和调节能力，导致生态失衡，并且影响感观的愉悦和水体的使用功能。图 5 所示是南湾湖中的鱼因缺氧而浮到水面进行呼吸。

以南湾湖为例,水草已经成为影响南湾湖景观质量及旅游活动的主要因子之一。每年农历5月初5端午节,南湾湖Ⅱ湖都举办国际龙舟竞赛,这已经成为一项有国际影响的传统赛事,也是澳门旅游业的一件大事。然而,生长茂盛的金鱼藻严重干扰了水体中船舶的航行。为此,市政厅每年要投入大量人力物力,清理湖中的水草。



图5 南湾湖Ⅱ湖中的鱼因缺氧而浮到水面进行呼吸

4. 垃圾污染

除了上述问题以外,南湾湖水体也受到垃圾的污染。这些垃圾主要来源于游人,也有一些是来自周边建筑工程施工的建筑垃圾。这些垃圾也严重影响了南湾湖作为旅游环境的价值。



图6 南湾湖Ⅱ湖中漂浮生活垃圾

5. 初步结论与建议

根据以上分析,南湾湖定位为非人体直接接触的景观娱乐水体,与此不相适应的是,该湖泊已经受到(1)水质污染;(2)富营养化;(3)水草过盛;(4)垃圾污染;等问题的威胁。

上述问题又与南湾湖的管理体制有关，为此，笔者提出以下建议：

- (1) 加强对南湾湖的监测和监督管理；
- (2) 对污染源进行细致调查，从源头削减污染物的排放；
- (3) 针对水草问题开展专项研究，寻找适宜的控制对策；
- (4) 加强制度建设，制定南湾湖水环境管理办法；
- (5) 开展规划研究，因地制宜地确定控制目标和管理办法。

Environmental Problems and its Countermeasures of Nam Van Lake, Macao

P. Du, S. Zeng, J. Chen

(Environmental Simulation and Pollution Control State Key Joint Laboratory, Department of Environmental Science and Engineering, Tsinghua University, Beijing 100084)

Wang Zhishi, Tang U Wa

Faculty of Science and Technology, University of Macao, Macao

Abstract: According to long period monitoring and field investigating, the main environmental problems of Nam Van Lake are as follow: i. Water quality pollution; ii. Eutrophication; iii. Float grass explode; iv. Rubbish pollution and landscape derogatory; v. weakness of environmental management. To solve these problems, some countermeasures are given here:

- i. enhancement of monitoring and supervising;
- ii. investigation and control of pollution sources, ;
- iii. approach on float grass and seek feasible control methods;
- iv. management system construction;
- v. make environmental protection planning.

Keywords: Nam Van Lake; Eutrophication; Float Grass; Environmental Management

澳门南湾湖的底泥污染分析*

冉圣宏 杜鹏飞 曾思育 陈吉宁

(清华大学环境科学与工程系, 环境模拟与污染控制国家重点联合实验室, 北京 100084)

王志石, 邓宇华

(澳门大学科技学院, 澳门)

摘要: 南湾湖是澳门本岛的一个人工湖, 未来几年它将成为澳门的一个旅游观光热点, 但目前该湖已经面临湖泊水质变差、富营养化等一系列问题。对南湾湖水水质污染物的来源及其污染机理的分析表明, 底泥中污染物的重新释放是影响南湾湖水质的一个重要因素: 历史上南湾湖是一个海湾, 澳门本岛的污水长期排放到这里, 这些污染物中一部分已经降解, 而另一部分沉积到底泥中, 在适当的条件下可能会溶出而重新进入水体, 使得水体中该污染物的浓度偏高, 如总磷浓度。因此对南湾湖的底泥进行评价是提出南湾湖生态环境综合整治措施的基础之一。

关键词: 南湾湖, 底泥污染, 富营养化

对南湾湖水水质污染物的来源及其污染机理的分析表明, 底泥中污染物的重新释放是影响南湾湖水质的一个重要因素: 历史上南湾湖是一个海湾, 澳门本岛的污水长期排放到这里, 这些污染物中一部分已经降解, 而另一部分沉积到底泥中, 在适当的条件下可能会溶出而重新进入水体, 使得水体中该污染物的浓度偏高, 如总磷浓度。因此对南湾湖的底泥进行评价是提出南湾湖生态环境综合整治措施的基础之一。

1. 底泥评价的依据与方法

为了准确地对南湾湖底泥污染状况进行评价, 必须有恰当的评价标准。但目前国家颁布的环境质量标准中尚无湖泊底泥类标准, 较常用的评价方法是以当地湖区土壤中相应污染物的自然含量(或背景值)加上两倍标准差作为标准进行评价。也有一些研究将国家地表水 III 类标准进行修订后作为底泥评价标准, 或经过毒理实验确定相应的评价标准。由于各种原因, 本文没有对澳门南湾湖底泥元素进行毒理实验, 仅对南湾湖底泥质量作一定性分析, 并对其时间与空间的分异情况进行分析。在评价过程中的主要依据是:

- 《中国土壤元素背景值》
- 《中国土壤》
- 《中国土壤肥力》
- 《广东土壤》
- 《海洋沿岸底质污染物标准》
- 《土壤环境标准》

2. 底泥评价的数据来源及评价依据

为了对南湾湖水水质及其底泥进行评价, 澳门市政厅专门组织了几次监测, 于 2000 年 2 月对南湾湖表层底泥进行了分析, 2000 年 4 月—2000 年 7 月对南湾湖水体水质的多项指标进行

* 本研究得到澳门市政厅及澳门大学的资助。

了监测，2000 年 12 月又委托中科院地球化学研究所（广州）分别对 I 湖（东）和 II 湖（西）的底泥取柱样进行了分析，本文就是在上述工作的基础上对南湾湖底泥的污染状况进行评价的。

根据《中国土壤》、《中国土壤元素背景值》以及《广东土壤》等从土壤类型、行政分区等角度所做的广泛调查，广东省北纬 21°35′~24°31′之间主要分布赤红壤类花岗岩赤红壤亚类土壤、盐渍水稻土以及滨海盐土类滨海沼泽盐土亚类等，广东珠江三角洲和澳门就属于该地区，对调查结果进行分析总结，可知广东珠江三角洲地区土壤的主要理化性质，见表 3-1。澳门南湾湖土壤中各元素的背景值即以此为主要依据。

表 1 广东省珠江三角洲土壤中锰元素和有机质的背景值

土层深度(cm)	有机质(%)	全氮(%)	全磷(%)	速效磷(ppm)	锰 (mg/kg)
0~16	2.63~3.73	0.146~0.164	0.042~0.186	5.3~18	855 (1135)

3. 南湾湖底泥总体评价结论

3.1. 总体评价

表 3-2 列出了南湾湖底泥 2000 年 2 月的监测数据。由于当时没有监测全氮指标（而是氨氮指标），因此用 2000 年 12 月 10 日监测的全氮指标代替。表中最后两行是 M01、M02 两个柱样的分析数据，列在这里是用来与其他数据进行比较，说明两次监测方法的差别。

表 3-2 南湾湖底泥监测数据*

编号	pH	总磷 (P ₂ O ₅ , mg/kg)	总锰 (Mn, mg/kg)	全氮 (g/kg, 干重)	总氮 (NH ₃ -N, mg/kg)	有机物(%)
M1-1	6.4	2070	583	2.183 (M22-1)	10	2.1
M1-2	7.4	919	760	2.258 (M51-1)	11	1.3
M1-3	7.7	1660	684	1.659 (M17-1)	10	0.8
M1-4	7.5	2080	569	2.134 (M18-1)	14	1.2
M2-1	7	2250	633	2.616 (M25-1)	14	1.9
M2-2	7.6	1330	598	2.189 (M52-1)	4.2	0.18
M2-4	7.4	2250	417	1.585 (M53-1)	5.9	1.1
M2-6~M2-7	7.5	1670	621	1.502 (M61-1)	8.5	0.78
M01柱样		1230		4.788		
M02柱样		917		3.343		

注：1 全氮指标是 2000 年 12 月 10 日的监测数据，该列括弧中列出的是监测点位置，与 2000 年 2 月的监测点位有一定差别。

2 两个柱样分别取自 2000 年 12 月 10 和 2000 年 12 月 11 日，M01 位于 I 湖东北部，M02 位于 II 湖中心。

从以上监测数据可以看出，底泥中总磷的污染相当严重，平均值达到 1778.6mg/kg，极大值为 2250mg/kg，远远超过其背景值 700mg/kg；全氮含量的平均值达到 2.016g/kg，极大值为 4.788 g/kg，也已超过了其背景值 1.46~1.64 g/kg 的范围，但污染程度比总磷略轻；锰的含量与土壤背景值（约 500~1000mg/kg）相当，初步认为南湾湖底泥中的锰含量受人类活动的影响不大。

有机质含量也没有明显异常，与一般底泥中有机质的背景含量 10g/kg（1.0%）相当，但从仅有的 8 个监测点来看，它具有一定的空间分异规律，南湾湖西部（II 湖）的几个测点略高，

可能受到了人为活动的影响。

3.2. 底泥中氮磷含量的空间分布

2000 年 12 月 10 日对南湾湖底泥的表层进行了多个采样，并对样本中的全氮、全磷以及速效氮的含量进行了分析，如图 1。从图中可以看出，无论是总磷还是总氮，最大值与最小值的差别均在 1 倍左右，由于湖泊的总面积不到 0.6Km²，在这样小的区域范围内其总磷和总氮含量的差别如此之大，应认为南湾湖总磷和总氮的污染有明显的空间变化。

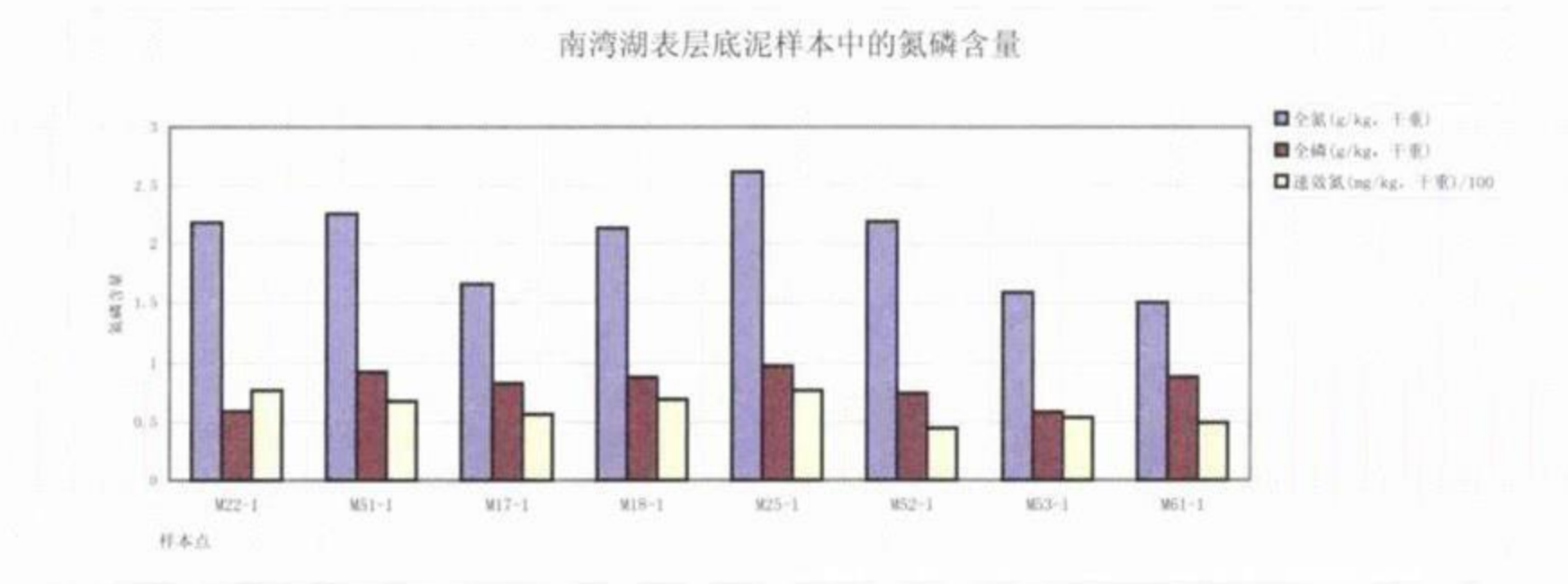


图 1 南湾湖表层底泥样本中氮磷含量分布

3.3. 底泥中氮磷含量的时间分布

根据对 2000 年 12 月 10 日和 11 日分别采取的两个柱样不同深度的氮磷含量进行分析，可以得到它们的时间变化趋势，并可据此进一步分析人为活动对湖泊底泥的影响。

从对 M01 柱样不同深度全氮含量的分析可以看出，全氮在 50 年代以前基本稳定在 2g/kg 左右，此后虽有较大的波动，上升趋势明显，目前已达到 4.788g/kg；在 M02 柱样中，出现异常值 21.706g/kg（60 年代），全氮含量在近年也有明显提高，为全氮背景值的 300 倍左右，表明近年来有含氮污染物排入。

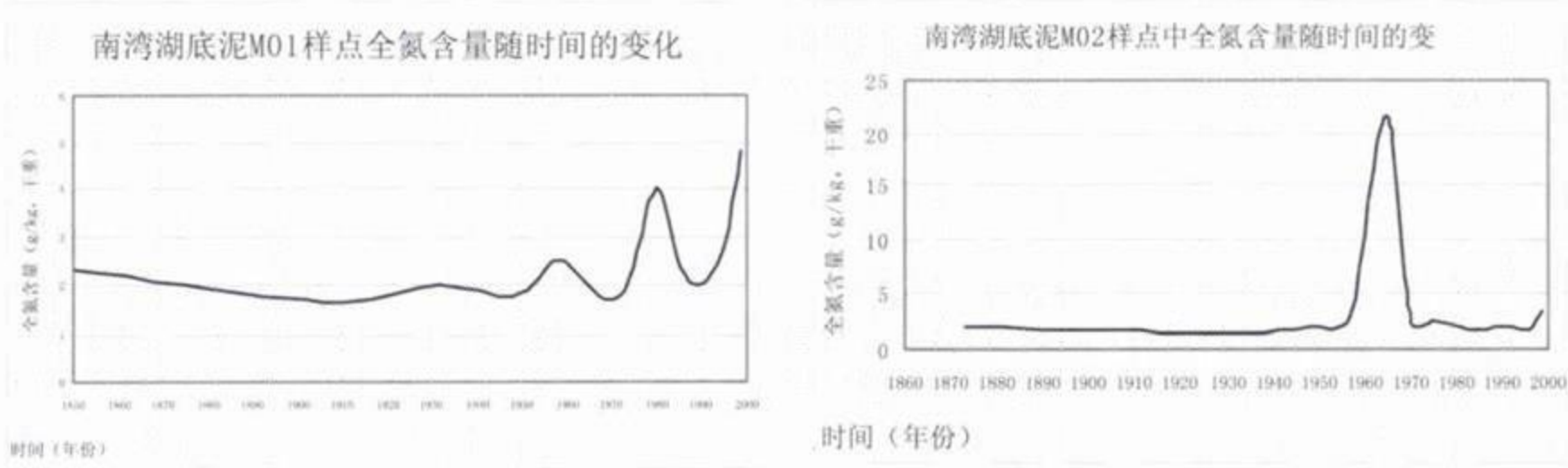


图 2 南湾湖 M01、M02 底泥样本中全氮随时间分布情况

对底泥全磷含量的分析表明，M01 柱样表明南湾湖底泥中全磷的含量在 90 年代中后期有较快的增长，目前全磷含量已达到 1.2g/kg；M02 柱样则显示自 30 年代起底泥中全磷含量即开始波动上升，目前基本稳定在 0.8g/kg 左右，是全磷含量背景值的 200~600 倍，底泥中磷的污染已经相当严重。

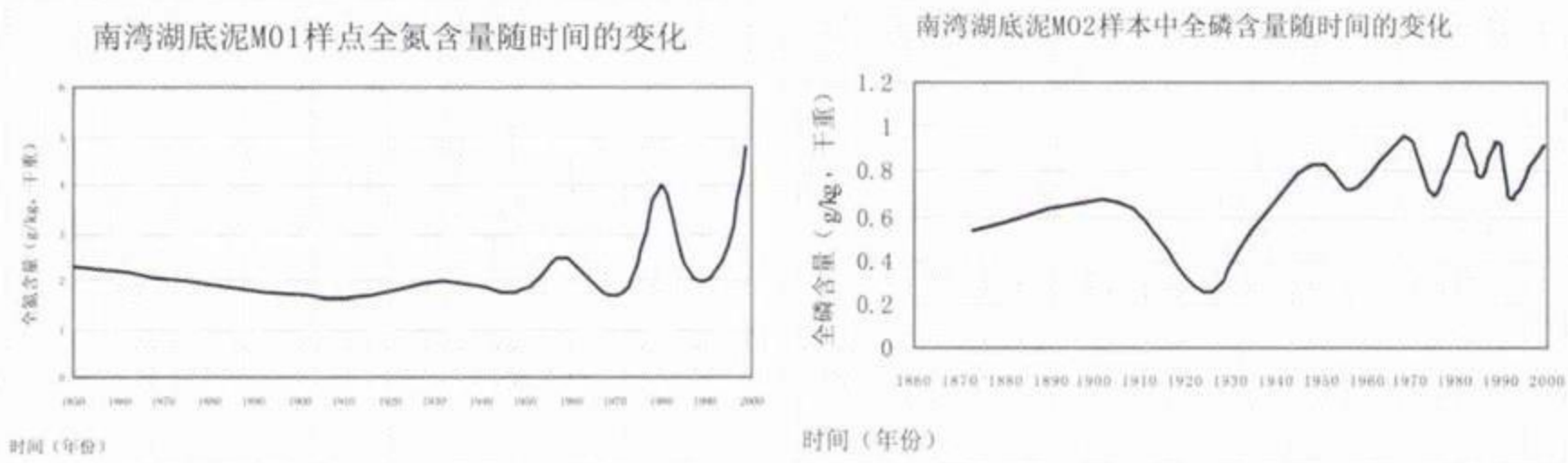


图 3 南湾湖 M01、M02 底泥样本中全氮随时间分布情况

对两个柱样中速效氮含量的分析也表明它们在近年有所上升，明显受到人为活动的影响，但上升趋势不如全氮和全磷明显，这与排入南湾湖的污染物的组成成分有关。

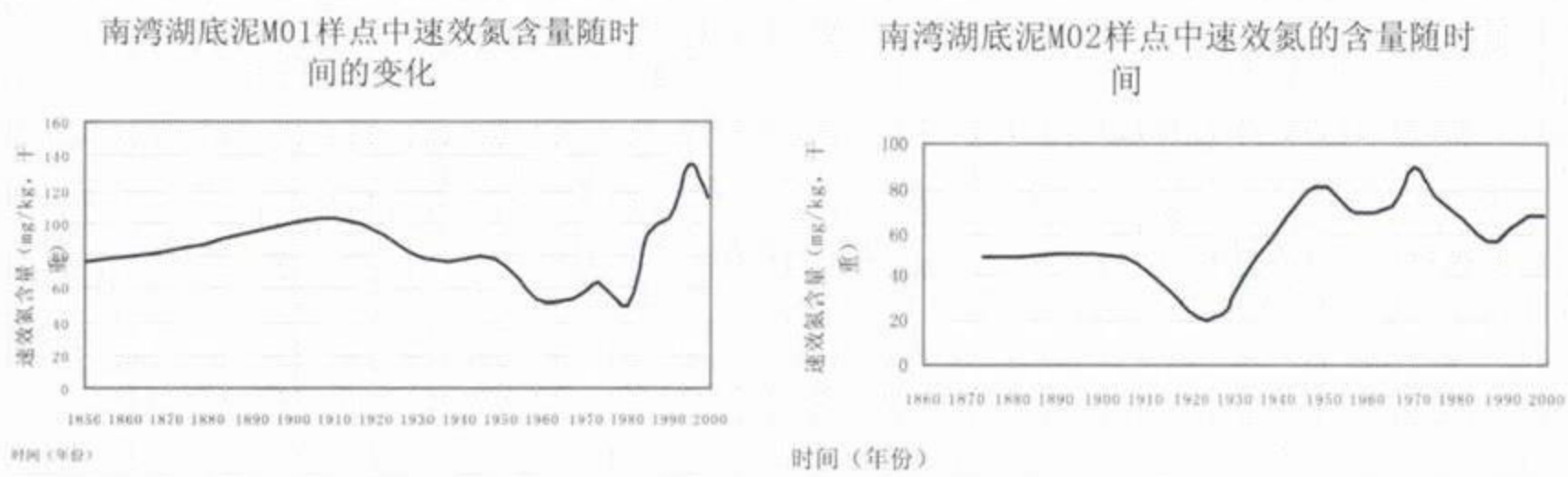


图 4 南湾湖 M01、M02 底泥样本中速效磷随时间分布情况

4. 初步结论

以上分析表明，南湾湖在开发初期污染严重，近几年仍有上升趋势，表明人类活动对底泥的影响较大。从总磷与总氮的时间变化趋势可以看出，人类活动对南湾湖的影响在近几年尤为明显，两个柱样的分析结果都表明总磷与总氮在 90 年代都有明显的增长；从总磷与总氮污染的空间差异也可以看出，湖泊的不同位置受到的影响不完全相同，总磷和总氮都在 M25-1 监测点达到最大值，最小值分别在点 M53-1（该监测点的总氮浓度仅略大于 M61-1 监测点）和 M61-1 取得，这种空间差异可能是由监测点位与排污口距离不同引起，再次表明人类活动的影响明显。

澳门大气有机污染物组成、来源与初步影响评价

王新明, 盛国英, 张志强, 傅家谟

(中国科学院广州地球化学研究所有机地球化学重点实验室)

王志石, 邓宇华

(澳门大学科技学院)

摘要: 通过对澳门地区空气中挥发有机物、颗粒物(TSP、PM₁₀)中多环芳烃的采样分析, 初步探讨了澳门地区大气有机污染物的组成与来源特征, 并进行了初步的影响评价。澳门地区空气中毒害挥发有机物主要为苯系物, 在街道微环境中的健康影响应引起注意。挥发有机物的来源中, 机动车尾气的贡献达 60%以上, 冬春季节在东北季风条件下珠江三角洲地区的大气输移可能对澳门地区大气挥发有机物也有较大贡献。与珠江三角洲地区其它城市相比较, 颗粒物中多环芳烃的含量处于较低水平, 其主要来源为以汽油为燃料的机动车排放。

关键词: 空气污染, 有机污染物, 澳门

空气中一些有机污染物会经吸入或皮肤吸收对健康带来直接危害(WHO, 1999)。空气中挥发有机物的大气光化学过程还会生成二次污染物(如臭氧和二次气溶胶)(Carter, 1990; Crosjean, 1992; Bowman, et al., 1995; Odum, et al., 1997), 对健康和生态带来严重影响。在澳门地区, 通过常规空气质量监测和其它一些研究, 对该地区二氧化硫、氮氧化物、一氧化碳等污染物的状况已有较深入的了解, 而对空气有机污染物的了解则相对缺乏。本文主要结合在澳门进行的数次的大气有机污染物采样, 对澳门地区大气有机污染物组成、来源以及其健康影响进行初步的分析。

1. 采样与实验分析

1.1. VOCs 采样与分析

挥发性有机物分析方法主要为吸附管吸附-热脱附法。捕集管购自美国 Tekmar 公司, 内部充填有 Tenax 和碳分子筛。采样时采用江苏建湖电子仪器厂生产的 THW-1500 大气采样器进行采样。分别于 1995 年 11 月和 2000 年 12 月于澳门半岛典型街道进行了采样, 采样时采样高度均为距地面 1.2 米。2000 年 12 月还在澳门大学何楼(Stanley Ho Building)楼顶进行了连续采样, 在大潭山进行了采样, 同时还针对一些 VOCs 排放源进行了采样分析。2001 年 5 月用采样罐采集了一些样品。

野外采样后的捕集管安装到 Tekmar 6032 Aerotrap 的热脱附位置, 快速升到高温进行热脱附, 脱附产物被氦气流带到 Tekmar 3000 Purge & Trap Concentrator 经由捕集阱被捕集起来, 脱附过程完成后, 捕集阱快速升至高温(升温速率 $>400^{\circ}\text{C}/\text{min}$), 氦气流将捕集阱脱附出来的有机化合物带到 HP 5972 GC/MSD 系统进行定性定量分析。采样罐采集的样品用 Entech 7100 进行预浓缩后传输到 HP 5972 GC/MSD 系统进行定性定量分析。化合物定性主要依据质谱图及相对保留时间来, 采用标样用静态外标法定量。

1.2. 颗粒物采样与分析

颗粒物采样分别在 1995 年 11 月、1998 年 11 月、2000 年 12 月进行了采样, 其中 1995 年

和 1998 年只进行了 TSP 采样,2000 年除 TSP 外还采集了 PM10 样品。TSP 采样用国产采样器,采样流量为 1050L/min,样品收集在 8"×11"石英滤膜上。PM10 采样用 Anderson 采样器,流量分 16.7L/min,样品收集在直径为 47mm 的玻璃纤维滤膜上。

收集在滤膜上的样品以二氯甲烷为溶剂进行超声萃取,萃取液经浓缩和溶剂置换后通过硅胶柱分离出芳烃组份,芳烃组分在 HP 5972 GC/MSD 上进行定性定量分析[6]。

2. 结果与讨论

2.1. VOCs

2.1.1. 挥发有机物组成特征

用吸附管-热脱附法可检测出卤代烃和 C₅-C₁₂ 的烃类,用采样罐采样可分析卤代烃和 C₂-C₈ 的烃类。街道样品中绝大多数是用吸附管采样的,参照少量用采样罐采集的样品,可以初步得出澳门半岛街道样品中总体组成特征主要为: C₂-C₁₂ 烃类含量远大于卤代烃。其中 C₂-C₁₂ 烃类中有链烃和单环芳烃,一般是 C₂-C₄ 的链烃和苯系物含量较高。卤代烃中主要是氟氯烃、氯代甲烷类、氯代乙烯类和氯代苯类,一般是三氯乙烯和四氯乙烯含量最高。因此从直接健康影响来说,苯系物和氯代乙烯类化合物是街道空气中需重点关注的 VOCs。

从 1995 年与 2000 年的比较来看(图 1),几种主要毒害性挥发有机物中,2000 年甲苯含量升高,可能与无铅汽油使用或溶剂排放增加有关;其它几种浓度均有所降低,特别是三氯乙烯和四氯乙烯降幅较大。

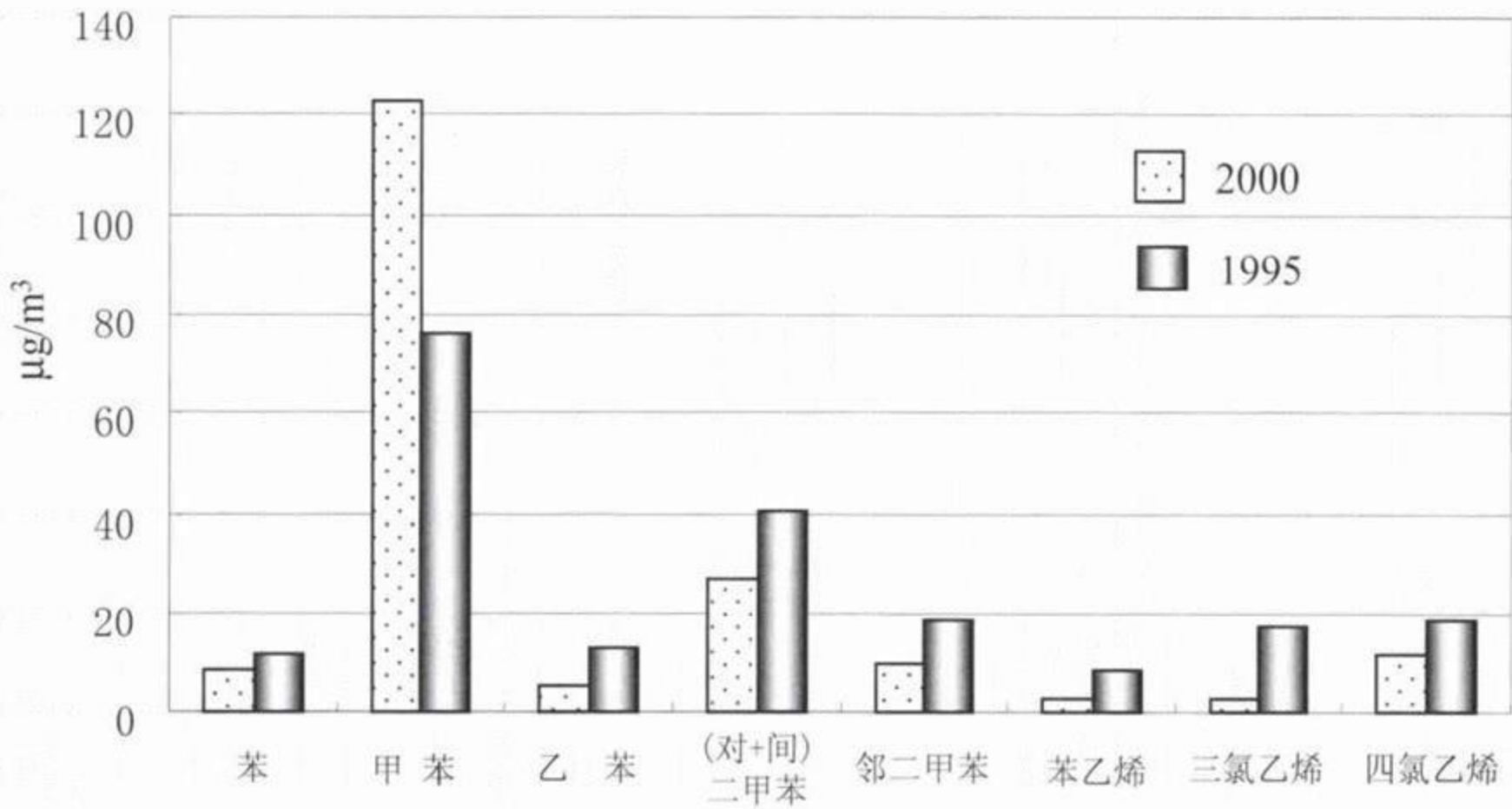


图 1 街道空气中几种主要毒害性 VOCs 比较 (2000 VS 1995)

与珠江三角洲其它城市相比较,1995-1996 年澳门和广州市区均具有高人口密度、高机动车流量的特征,相对于城市布局比较分散的南海,不仅单位长度街道机动车排放强度要高出很多,而且因高楼林立使得峡谷效应更加明显。图 2 是澳门与广州和南海街道苯系物平均含量与变动范围的比较,可以看澳门的苯含量低于广州,其它几种则略高于广州,但比南海均高出很多。表 1 是 2000 年 12 月澳门半岛与广州市中心街道空气中几种主要毒害 VOCs 比较,可见苯系物中澳门苯与乙苯含量相对于广州要低得多,二甲苯类与广州相当,但甲苯要比广州高。卤代烃中澳门三氯乙烯和四氯乙烯均比广州高,特别是四氯乙烯是广州的 5 倍多。

表 1 2000 年澳门半岛与广州市中心街道空气中几种主要毒害 VOCs 比较

化合物	澳门			广州		
	平均值	最大值	最小值	平均值	最大值	最小值
苯	8.5	22.6	1.8	40.6	171.8	11.8
甲 苯	122.4	512.3	12.3	71.9	383.9	10.8
乙 苯	5.4	22.3	0.2	15.5	70.0	1.1
(对+间) 二甲苯	27.0	76.1	0.8	29.4	189.4	1.8
邻二甲苯	9.8	33.2	0.7	10.5	77.4	0.5
苯 乙 烯	2.5	6.8	0.1	8.2	23.7	0.3
三氯乙烯	2.6	5.0	0.5	1.6	38.3	0.0
四氯乙烯	11.9	37.6	2.0	2.1	17.9	0.2

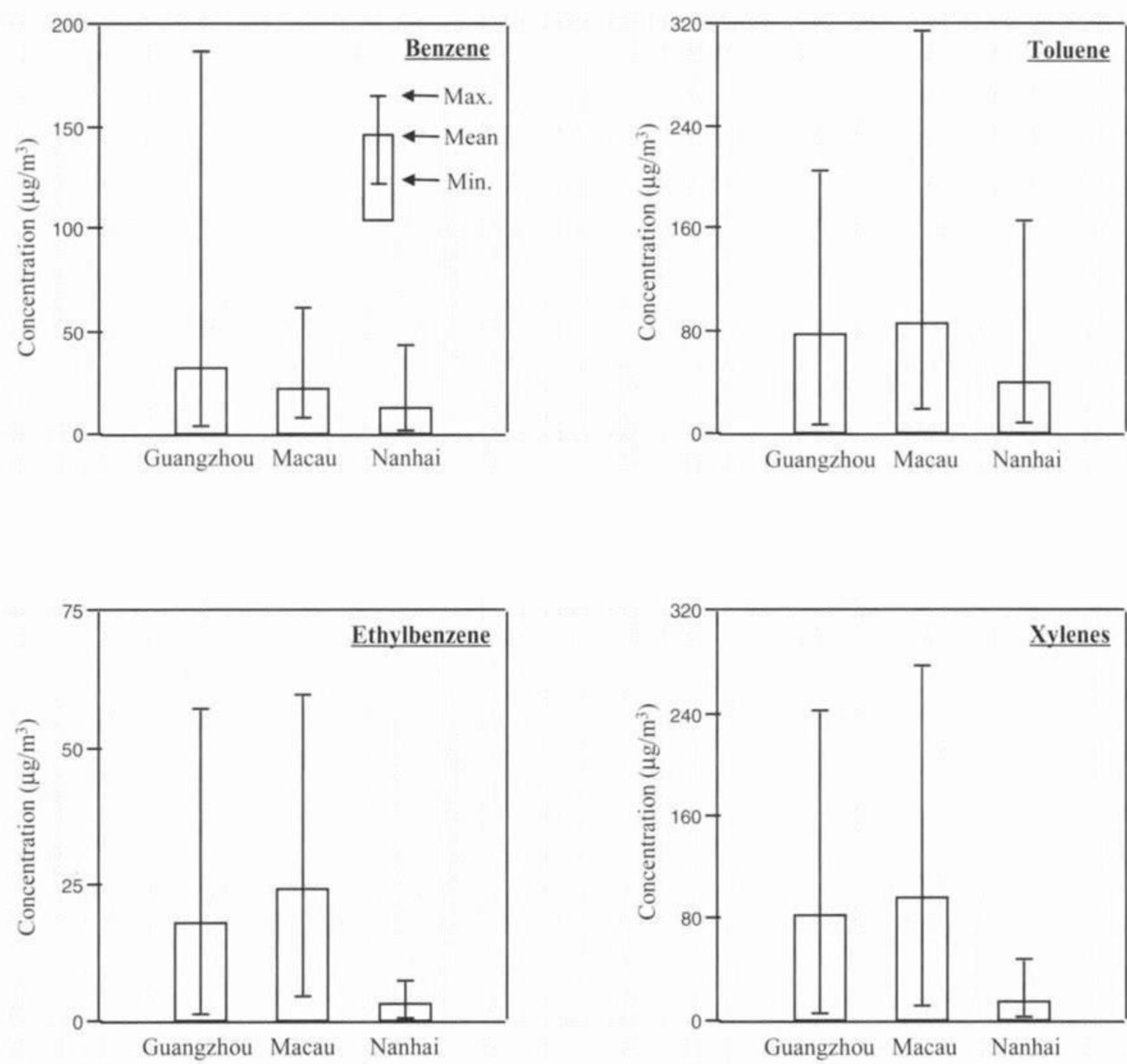


图 2 澳门（1995）与广州（1996）和南海（1996）街道苯系物含量比较

2.1.2. VOCs 来源特征

表 2 列出了澳门、广州和南海三市街道空气中 BTEX 相互之间的相关系数，可以看出三个城市中乙苯和二甲苯相互之间的相关系数均在 0.90 以上，表明其来源比较一致，主要是尾气来源；澳门甲苯与乙苯和二甲苯的相关系数同样在 0.9 以上，只是苯与其它几种的相互系数

在 0.8 附近，总体相互之间的相关系数均较高，表明其来源可能比较单一，以机动车尾气占绝对优势；广州苯与其它几种的相关系数均较差，表明其有异于机动车尾气的显著来源；而南海除乙苯和甲苯之间有很好的相关性以外，相互之间的相关性均较差，表明其来源的复杂与多样性。对于一些氯代烃类，因澳门没有重工业或化工业，作为溶剂或清洗剂成份是其

表 2 街道空气中苯系物相互之间的相关系数 (r)

化合物	广州				澳门				南海			
	B	T	E	X	B	T	E	X	B	T	E	X
B	1	0.15	0.29	0.21	1	0.77	0.82	0.84	1	0.06	0.27	0.22
T		1	0.76	0.94		1	0.97	0.92		1	0.39	0.45
E			1	0.91			1	0.96			1	0.94
X				1				1				1

B: 苯; T: 甲苯; E: 乙苯; X: 二甲苯

区内这些化合物进入环境的主要途径，当然不能排除澳门以外的珠江三角洲地区的输入。对于澳门街道空气中毒害性 VOCs，无论是 1995 年还是 2000 年，通过一定的模式进行源解析均表明机动车尾气的贡献不少于 60%.

2.1.3. 异地输入对区内 VOCs 的贡献

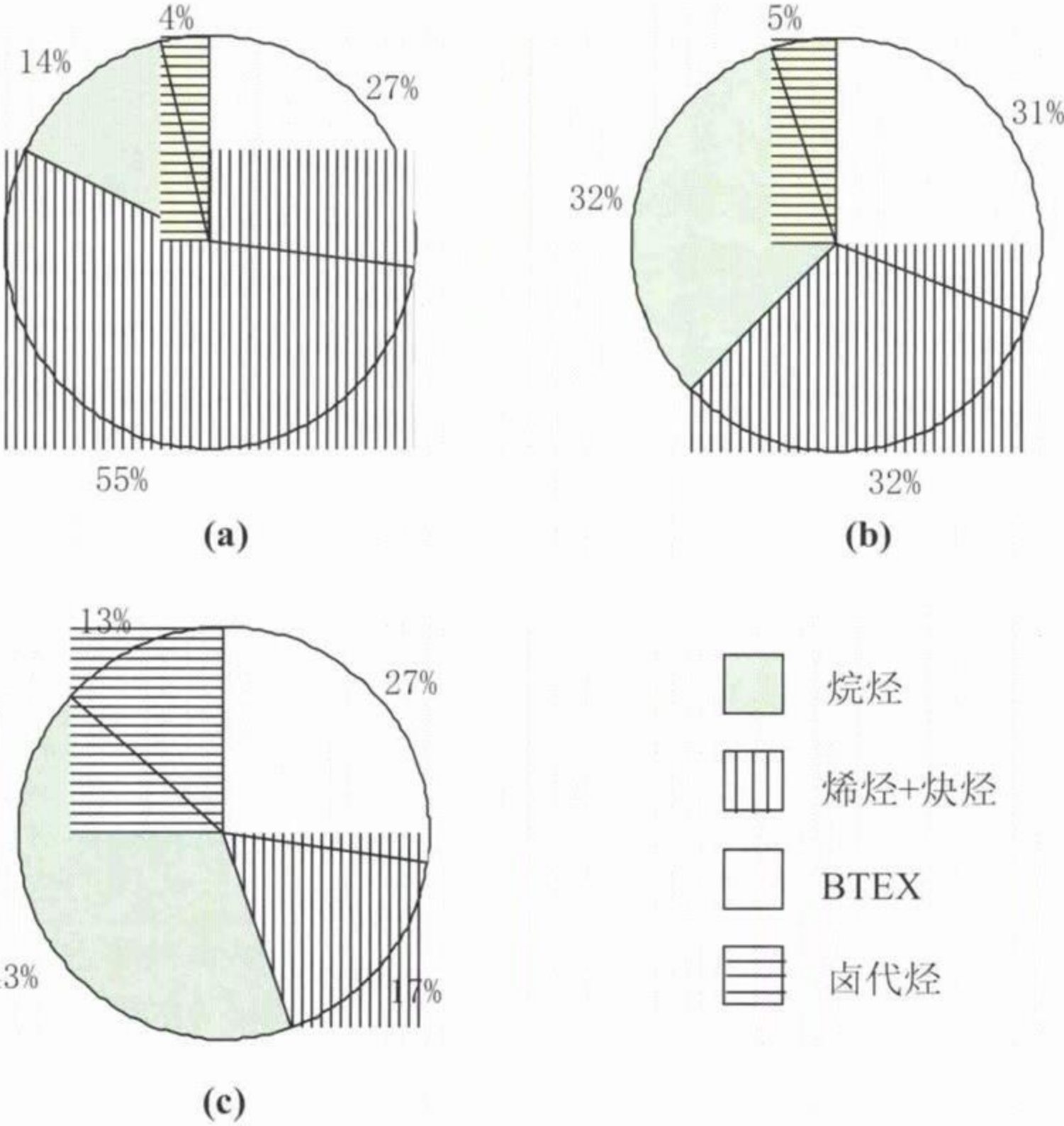


图 3 C2-C8 烃类和卤代烃组成的变化：(a) 半岛街道；(b) 澳大何楼；(c) 大潭山。

图 3 是澳门半岛街道空气样、澳门大学何楼 (Stanley Ho Building) 楼顶以及大潭山 VOCs 所表明出的组成特征。可以明显看出，澳大何楼和大潭山样品中反应活性强的不饱和烃类所

占比例已明显减少,而烷烃和卤代烃所占的比例相应增加,由于澳门地区绝大部分人类活动集中在澳门半岛,而机动车尾气是该地区的主要挥发有机物排放源,由于澳大和大潭山与澳门半岛之间的距离较近,结合气象因素,在此距离内大气光化学过程不足以组成上如此大的悬殊。一个合理的解释是,街道空气中挥发有机物因峡谷效应主要与机动车尾气有关,而澳大何楼楼顶与大潭山因地势较高,其挥发有机物不仅有澳门本地的贡献,相当比例可能与珠江三角洲腹地排放的挥发有机物的大气运移有关。何楼楼顶和大潭山样品中苯/甲苯比值(分别为0.13和0.21)比街道样品高(0.07),由于甲苯的光化学速率远比苯快,所以在大气输运过程中苯/甲苯比例升高,仅在澳门半岛和氹仔岛之间的大气光化学过程不足以造成这种比例的差异,何楼样品中较高的苯/甲苯比值同样表明有比较明显的异地输入。

2.2. 颗粒物中的多环芳烃

2.2.1. 16种优控多环芳烃的含量情况

1995年和1998年采样主要集要中澳门半岛,2000年样品主要在气象台和澳门大学。表3和表4列出了1995年和1998年16种优控多环芳烃的含量情况。1995年所测得的16种优控多环芳烃总量 Σ PAHs为9.0-38.6ng/m³,1998年 Σ PAHs范围为10-51ng/m³(夜间)和11-80ng/m³(白天)。Cotham et al.(1995)报道的美国Denver、Portland、Columbia和Chicago的含量分别为93ng/m³、57ng/m³、43-56ng/m³和75-1410ng/m³。因此澳门大气中多环芳烃处在一种相对较低的水平。在气象台所采的样品中总量则在10ng/m³以下。

表3 1995年月11月样品中16种优控多环芳烃的含量情况

Samples	M-95-1	M-95-2	M-95-3	M-93-4	M-95-5	M-95-6
Nap	0.75	n.d.	n.d.	n.d.	n.d.	n.d.
Ace	n.d.	n.d.	tr.	n.d.	tr.	n.d.
Dih	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Flu	0.06	n.d.	0.04	n.d.	0.4	n.d.
Phe	2.05	1.03	0.83	0.91	1.15	0.89
Ant	0.21	tr.	tr.	tr.	tr.	tr.
Flua	3.01	1.19	1.12	1.34	1.25	1.25
Pyr	4.34	2.19	1.19	1.5	1.26	1.15
Ben(a)	2.31	2.42	0.67	0.48	0.52	0.4
Chr	3.39	2.8	1.24	0.86	0.83	1.94
Ben(b)	3.82	2.49	1.91	n.d.	1.04	1.48
Ben(k)	4.17	n.d.	1.51	n.d.	0.76	2.13
Ben(a)p	2.61	0.94	0.8	n.d.	0.48	0.52
Ind	4.21	2.8	2.01	1.09	0.83	1.29
Dib	0.46	0.51	tr.	tr.	tr.	tr.
Ben(ghi)	7.24	4.1	2.67	2.83	1.04	1.62
Σ Priority PAHs	38.63	20.47	13.99	9.01	9.56	12.67

Nap=Naphthalene; Ace=Acenaphthylene; Dih=Dihydroacenaphthylene;Flu=Fluorene; Phe=Phenanthrene; Ant=Anthracene;Flua=Fluoranthene;Pyr=Pyrene;Ben(a)=Benzo(a)anthracene;Chy=Chrysene; Ben(b)=Benzo(b)fluoranthene;Ben(k)=Benzo(k)fluoranthene;Ben(a)p=Benzo(a)pyrene; Ind=Indeno(1.2.3.-cd)pyrene; Dib=Dibenzo(a,h)anthene; Ben(ghi)=Benzo(ghi)perylene. tr.=trace; n.d.=not determined * Total concentration of Ben(b) and Ben(k).

澳门大气气溶胶中多环芳烃的分布和广州荔湾区较为相似,二者均是贫2+3环,富4+5+6环;二环、三环的萘、苊、二氢苊、芴、蒽在TSP样品含量极低,其在PM₁₀中也极少检出,但具有三个环的菲在大多数样品中均有检出;所有样品中的多环芳烃主要是四环至六环的10种致癌或有毒理影响的化合物,其含量占总PAHs总量的90—95%。在TSP样品中还存在多

环芳烃化合物的含量随分子量增加而增大的趋势（如图 4（a,b））。其原因可能有二：① 由于澳门地处亚热带，常年的平均气温较高，导致挥发性 PAHs（即轻组分）在气相/颗粒相的比例加大，从而使其在颗粒物上的含量减少。②由于澳门没有工业，主要污染源为以汽油为燃料的机动车排放的废气。汽油车燃烧产物中 16 种优控多环芳烃均有检出，其中轻组分的相对含量较低,这与澳门大气气溶胶中多环芳烃的分布较为接近，而且 A.H.Miguel 也认为，柴油载重车是较轻多环芳烃的主要来源，而轻载汽油机动车则对高分子量 PAH 的贡献较大。所以以汽油为燃料的机动车可能为为澳门的主要污染源。

表 4 1998 年 11 月样品中 16 种优控多环芳烃的含量情况

PAHs	M-98-1		M-98-2		M-98-3		M-98-4		M-98-5	
	night	day	night	Day	night	day	night	day	night	Day
Naphthalene	tr.	tr.	n.d.	Tr.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Acenaphthylene	0.03	0.06	0.03	0.03	tr.	tr.	n.d.	n.d.	n.d.	n.d.
Acenaphthene	tr.	tr.	tr.	Tr.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Fluorene	0.11	0.17	0.08	0.17	0.03	0.06	tr.	0.05	0.03	tr.
Phenanthrene	0.82	1.45	0.74	1.35	0.24	0.57	0.21	0.16	0.19	0.24
Anthracene	0.16	0.25	0.11	0.17	0.03	0.03	0.00	0.00	0.00	0.00
Fluoranthene	1.54	2.84	1.51	2.62	0.53	1.04	0.48	0.42	0.45	0.59
Pyrene	3.06	5.65	2.75	4.67	0.71	1.52	0.69	0.69	0.64	0.88
Benzo[a]anthracene	2.93	6.48	2.49	4.01	0.58	1.10	0.42	0.53	0.32	0.35
Chrysene	4.92	10.51	4.55	7.40	1.48	3.13	1.32	1.22	0.90	1.17
Benzo[b]fluoranthene	4.87	8.87	4.65	7.57	1.67	3.60	1.59	1.91	1.27	1.31
Benzo[k]fluoranthene	4.15	5.90	4.44	4.89	1.67	2.84	1.59	1.49	1.17	1.25
Benzo[a]pyrene	5.29	7.90	4.79	4.92	1.16	1.96	0.95	0.85	0.74	0.75
Indeno[1,2,3-cd]pyrene	8.30	10.60	7.01	9.64	2.09	4.86	2.28	3.08	1.70	1.95
Dibenzo[a,h]anthracene	1.97	2.56	1.98	2.13	0.71	1.14	0.53	0.74	0.42	0.45
Benzo[g,h,i]perylene	12.87	17.08	9.62	15.22	2.78	6.66	2.86	4.83	1.93	2.35
ΣPAHs	51.01	80.31	44.73	64.78	13.69	28.51	12.91	15.97	9.75	11.27

澳门基本没有工业，它的污染源主要也是来自机动车的污染。肇庆鼎湖山自然保护区位于珠江三角洲经济区西北边缘，其北部、西部和西北部均为人口较为稀少的山地，自然植被覆盖较好，而对比澳门清洁区气象台和广东省肇庆鼎湖风景区的大气气溶胶中的多环芳烃数据，可以看出澳门较肇庆市污染程度要轻，这可能与珠江三角洲的本底较高有关，而由表 5 可以看出，肇庆鼎湖山大气气溶胶粒子中危害最大的当属 PM10（占总 TSP 的 80%），而澳门的细颗粒的污染不太严重，其 PM10 上吸附的多环芳烃含量仅占 TSP 总量的 36.5%。

表 5 肇庆与澳门的背景区中 TSP 与 PM10 中 PAHs 含量

样品编号	澳门		肇庆	
样品类型	TSP	PM10	TSP	PM10
采样时间	2000.12	2000.12	2000.11	2000.11
Nap	0.01	0.01	n.d	n.d
Ace	0.01	nd	0.01	n.d
Dih	nd	nd	n.d	n.d
Flu	0.01	0.01	0.01	n.d
Phe	0.10	0.05	0.15	n.d
Ant	0.03	0.03	n.d	n.d
Flua	0.22	0.05	0.36	0.39
Pyr	0.24	0.07	0.35	0.39
Ben(a)	0.05	0.04	0.11	0.13
Chr	0.31	0.20	0.56	0.49
Ben(b+j+k)	0.39	0.11	1.52	1.04
Ben(a)p	0.22	0.13	0.61	0.48
Ind	0.07	0.07	0.89	0.79
Dib	0.03	nd	0.54	0.50
Ben(ghi)	0.61	0.08	1.16	0.82
16 种多环芳烃总量	2.88	1.05	6.28	5.02
PM10 _{pah} /TSP _{pah}	0.365		0.80	

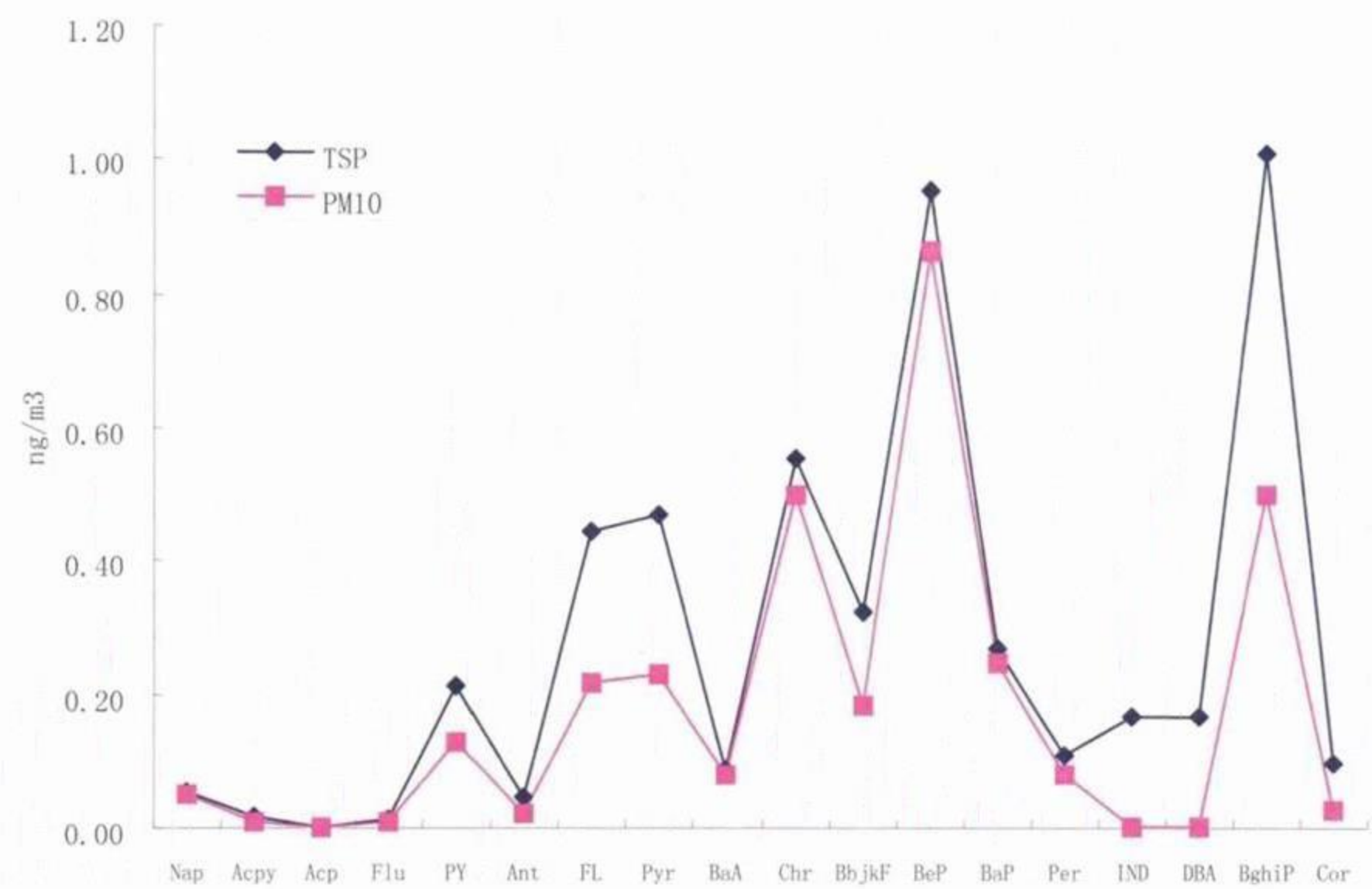


图 4 (a) 污水处理厂大气气溶胶中各粒级中 PAHs 分配



图 4 (b) 气象台大气气溶胶中各粒级中 PAHs 分配

2.2.2. 多环芳烃的来源分析

Grimmer 等人认为 IND/(IND+BghiP)的值为 0.18, 0.37, 0.56 时分别代表汽油, 柴油 和煤的燃烧。由上表可知, 气象台和污水处理厂二者的比值均在 0.18 附近, 表明其主要的污染源仍来自于以汽油为燃料的机动车污染。FL/(FL+Pyr)的值 0.44-0.53 之间, 同样表明了污染源主

要来自汽车尾气 (Rogge et al., 1993) (见表 6)。BeP/(BeP+BaP)的值在 0.55-0.61 之间, 表明污染物主要来源于汽油燃烧的排放物。

表 6 气象台与污水处理厂附近大气气溶胶中多环芳烃的特征比值

	气象台	污水处理厂
BeP/(BaP+BeP)	0.69	0.74
IND/(IND+BPe)	0.10	0.13
FL/(FL+Pyr)	0.47	0.47

2.3. 初步风险评价

以上的分析表明, 澳门地区大气颗粒物中多环芳烃含量水平处于较低水平, 对健康影响相对较小。街道环境中挥发有机物在不能有效扩散的情况下易出现较高浓度, 从而导致不利的健康影响, 其中苯系物应引起注意。WHO (1999) 统计指出, 空气中苯浓度每增加 1µg/m³, 每百万人中癌症患者会增加 4-8 人。欧洲一些国家建议室外大气中苯的浓度控制在 10µg/m³ 以下, 澳门街道空气中苯浓度超过 101µg/m³ 还相对比较普遍。另外, 整个珠江三角洲大背景对澳门地区毒害有机污染物和二次污染物生成的影响和贡献, 应进行较深入地探讨, 以有利于科学的污染控制决策。

3. 主要结论

澳门地区空气中毒害挥发有机物主要为苯系物, 在街道微环境中的健康影响应引起注意。挥发有机物的来源中, 机动车尾气的贡献达 60%以上, 但珠江三角洲地区的大气输移可能对澳门地区大气挥发有机物也有较大贡献。与珠江三角洲地区其它城市相比较, 颗粒物中多环芳烃的含量处于较低水平, 其主要来源为以汽油为燃料的机动车排放。

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Composition, Source And Preliminary Risk Assessment of Airborne Organic Pollutants in Macau

Xinming Wang, Guoying Sheng, Zhiqiang Zhang, Jiamo Fu

(State Key Lab. Of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640)

Zhishi Wang, U Wa Tang

(Faculty of Science & Technology, University of Macau)

Abstract: Volatile organic compounds (VOCs) as well as polycyclic aromatic hydrocarbons (PAHs) in particulates (TSP and PM₁₀) were sampled and analyzed in Macau for a primary investigation of the composition and sources of toxic organic pollutants in air, and for a preliminary assessment of their health risks. Monocyclic aromatic compounds revealed to be major toxic VOCs, and more than 60% of them were sourced from vehicle exhaust. In winter and spring VOCs transport from the inland of the Pearl River Delta might also contribute to the VOCs in Macau due to the NE monsoon. Levels of PAHs in Macau were relatively lower than those reported in other cities, and gasoline vehicles were found to be the major source of PAHs occurred in Macau air.

Keywords: air pollution; organic pollutants; Macau

Macao Noise Pollution Assessment and Control

Tam Vai Man* Zhishi Wang

Faculty of Science and Technology, University of Macau

Abstract: Through statistically analyzing data obtained from noise survey and regular monitoring, the acoustic environment in Macao peninsular is profiled. Traffic noise is the most important source of noise and its relation to environmental noise is qualitatively depicted. The result of using STAMSON (a computer model) to simulate environmental noise is quite satisfying. The average errors between simulation results and monitoring data are less than 2dB. Moreover, a brief but comparably comprehensive questionnaire was conducted to collect public response to noise exposure, especially on annoyance.

Key words: noise, acoustics, noise pollution, noise prediction, noise monitoring

1. INTRODUCTION

It has been demonstrated that community noise has direct adverse effects on human beings. They include adverse effects on communication, performance, behavior, noise-induced disturbance of sleep, community annoyance and so on. In Europe, almost 25% of the population is exposed to transportation noise over 65 dB(A) (24h Leq). When one realizes that at 65dBA sound pressure level, sleeping becomes seriously disturbed and most people become annoyed, it is clear that community noise is a genuine environmental health problem.

As a city that has a total area of 23.8 Km² and a population of about 450 000, the acoustical environment in Macao is even more unsatisfying. The significant increase in the number of complaints over the past years show that, on one hand, there is a growing public dissatisfaction on noise problem in the community, on the other hand, the existing controls on environmental noise have not gone far enough to meet community expectations.

2. MACAO PENINSULAR NOISE SURVEY 2000

2.1. Noise Survey Procedures

A square mesh grid map (300mx300m /cell) of Macao peninsular is used for selecting the noise monitoring sites. The measuring sites are chosen in the center or near to the cell center. In addition, some sites are selected near to the Macao region center in the cells. These sites are anticipated to show typical daytime and nighttime noise levels at specific locations by short duration grab sampling. The daytime and nighttime samples were collected by 15 minutes of continuous monitoring.

2.2. Data Analysis

* present address: Instituto Para Os Assuntos Cívicos e Municipais

2.2.1. Daytime Leq

The daytime average Leq is 66.5dB(A) in Macao peninsular. Compare this figure to the P.R.C. National Standard of Environmental Noise of Urban Area, the noise environment in Macao is unmatched to all types of residential area. It only coincides with the roadside environmental noise limit of 70dB(A), which is the worst situation allowed in the standard.

The range of daytime Leq is from 55.3dB(A) to 77.8dB(A) and the variation is 22.5 dB(A). It reveals that noise environment in Macao peninsular varies a lot in different areas. As *Figure 1* shows, there are 14 cells whose daytime Leq values are more than 70dB(A). This indicates that about 30% of the Macao peninsular area is under an unfavorable noise environment.

No.47 and No.109 are the noisiest zones during daytime because they are close to the very busy roads in Macao. Besides cell No.47 and 109, there are 12 cells whose noise environment at

daytime is also not acceptable because they attain noise level over 70dB(A).

On the other hand, there are 18 cells whose daytime Leq values are not satisfactory even they matched with the P.R.C. National Standard. If the noise environment does not improve in the future, these 18 cells will become noisier and not suitable for residential use anymore. In these 18 cells, though traffic noise is the major noise source, both commercial as well as industrial noise are subjects that could not be neglected.

2.2.2. Nighttime Leq

In the P.R.C. National Standard of Environmental Noise of Urban Area, roadside noise limit at nighttime is 55dB(A). However, the nighttime average Leq in Macao peninsular is 57.0dB(A), and it is 2dB(A) above the standard limit. This illustrates that the overall nighttime noise environment is very unsatisfactory in Macao peninsular. On the other hand, there are 30 cells whose nighttime Leq values are above 55dB(A). It is the other evidence indicating that more than 60% of area at nighttime in Macao is very noisy.

The range of Leq at nighttime is from 47.2dB(A) to 70.3dB(A), and the variation shows that the noise environment varies a lot in different usage of the cells. No.47 is still the noisiest zone of all. Again, the traffic noise is blamed for causing the high level of noise. The noise environment of nighttime is shown in *Figure 2*.

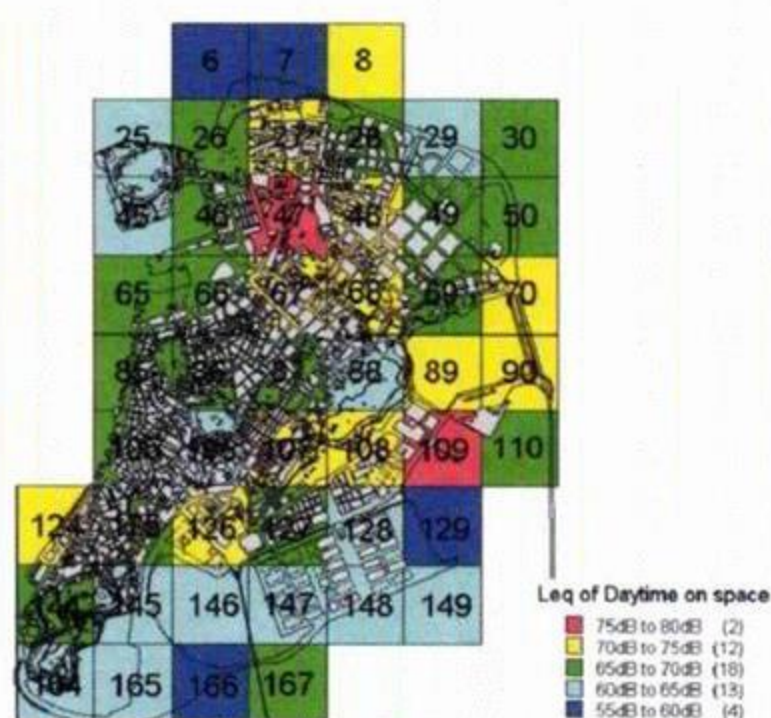


Figure 1 Daytime Leq on Space of Macao

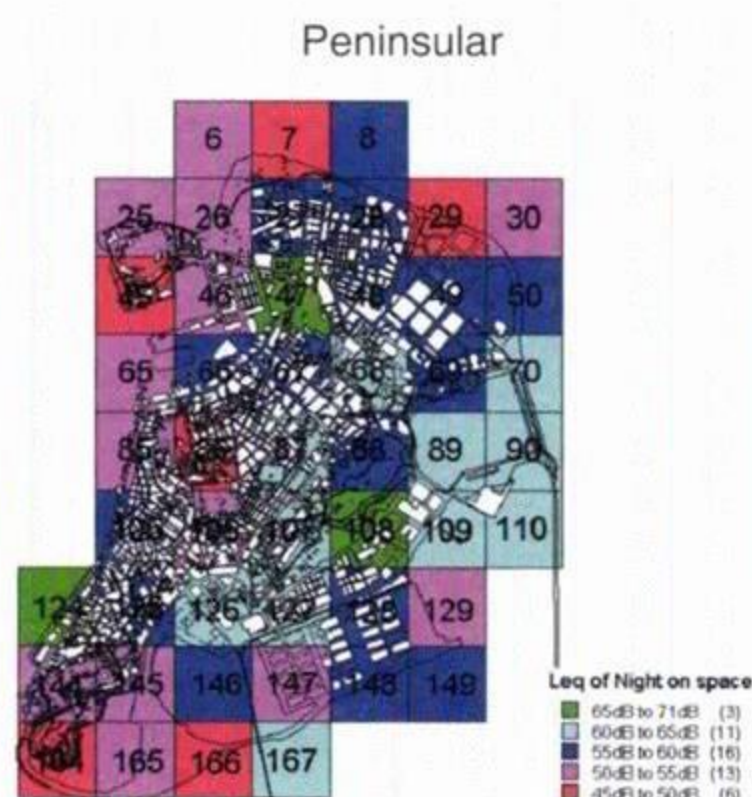


Figure 2 Nighttime Leq on Space of Macao

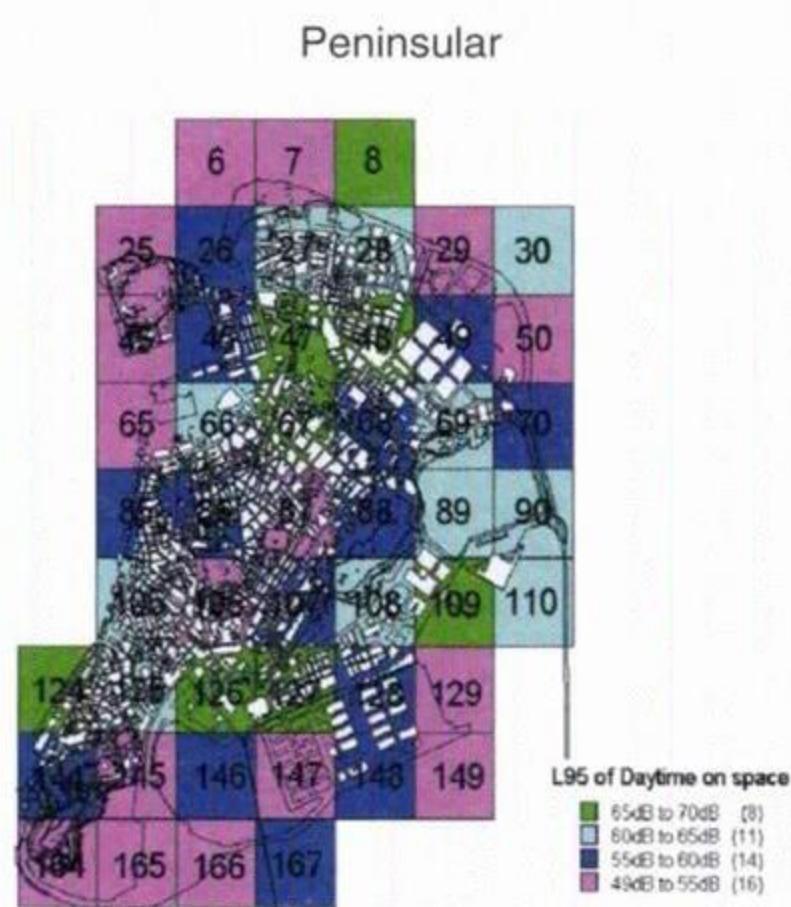


Figure 3 L95 of Daytime On Space

2.2.3. Daytime L95

L95 is used to represent the background noise level of a region. The average daytime L95 value is 58.2dB(A). This value is barely acceptable because it is around 60dB(A) that is considered as noisy situation in many western countries.

As Figure 3 shows, 16 cells are categorized in the group of 49-50dB(A) that means about 1/3 of the Macao peninsular area are under an acceptable noise environment. Except No.87 and No.106, the others are low residential density areas. A total of 30 cells whose daytime L95 is less than 60dB(A). This indicates that the noise environment in some 62% of Macao peninsular area is moderate, in spite of that fact that less than 40% of the population lives in those areas.

If the evaluation criterion changes to the legitimate reference of daytime background noise level - 65dB(A), the percentage will add up to 84%. That means most area in Macao is under this kind of noise environment. On the other hand, there are 8 cells whose daytime L95 levels are higher than 65dB(A). The major cause of high background noise level is the busy traffic flow, except No.8 and No.109 to which commercial activities are the other important noise sources.

Although the background noise situation is not satisfactory, there has not strict relation between background noise level and public nuisance. However, high background noise level can have bad effect on human health which should raise public concerns.

2.2.4. Daytime Difference Between Leq and L95

The difference between Leq and L95 represents the variation between environmental and background noise level. The greater the difference is, the more severe is the noise fluctuation and disturbances.

The average difference between daytime Leq and L95 is 8.4dB(A) that is slightly lower than the 10dB(A) legitimate limit. From this point of view, the legal standard may have some disadvantages in practical use because the actual noise environment almost exceeds it.

As Figure 4 shows, there are 17 cells or 35% of Macao peninsular area whose Leq and L95 difference are greater than 10dB(A). Even if traffic noise is always considered as liner noise source, its real characteristic is impulsive while the automobiles just come by the sound level meter in a short period of time. Moreover, since industrial noise and commercial noise are comparably steadier than traffic noise. A difference of 10dB(A) or more is an evident that traffic noise is the major environmental noise source.

23 cells are in the second category. As the traffic

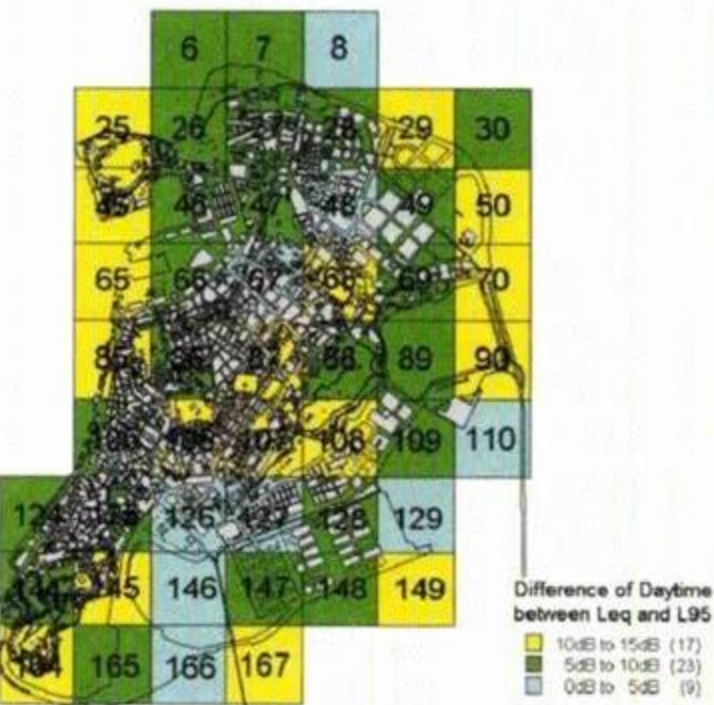


Figure 4 Daytime Differences Between Leq and L95

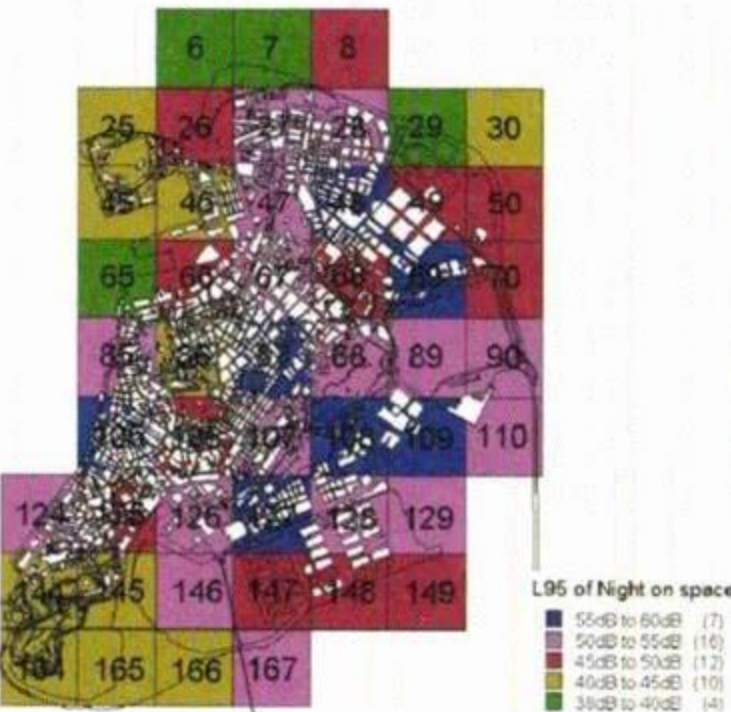


Figure 5 L95 of Nighttime On Space

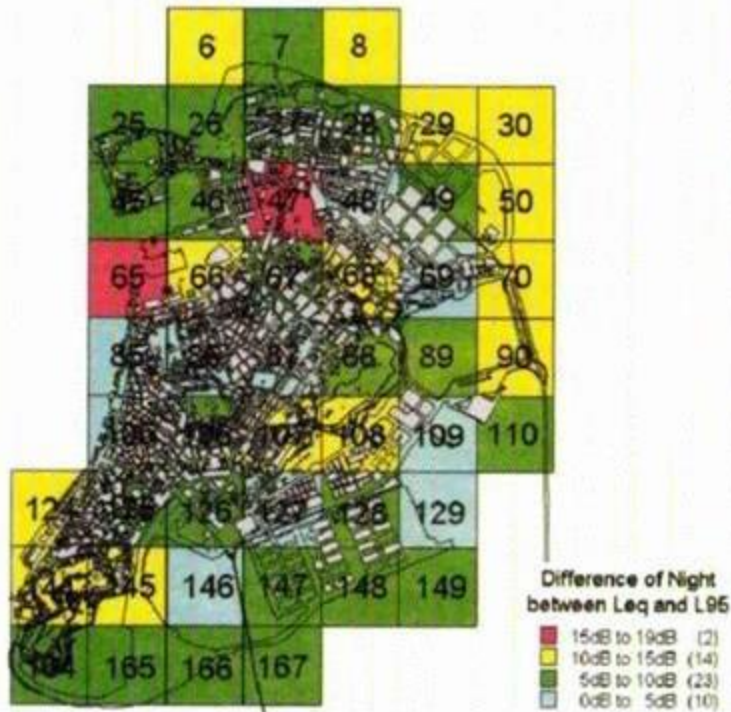


Figure 6 Nighttime Differences Between Leq and L95

volume is larger in these places, the amplitude of noise level fluctuation is less intense. Furthermore, 9 cells are categorized in the last group, 0dB(A) to 5dB(A). That means the noise environment is more stable in these regions. There are two possible explanations: (1) the traffic volume is so large that it diminishes the impulsive effect; (2) the traffic volume is not large enough to stir up noise fluctuation.

2.2.5. Nighttime L95

The legitimate reference of nighttime background noise level is 55dB(A). While using this criterion to evaluate the survey data, the outcome is satisfactory because only 7 cells exceed its standard and nearly 86% of the Macao peninsular area fulfills it. However, the average is 48.6dB(A) and is about 6dB(A) less than the reference. What is more, there are 26 cells whose nighttime background noise level is less than 50dB(A), and these cells cover about 53% of Macao area. So in practical use, the reference may be inadequate. (Figure 5)

2.2.6. Nighttime Difference Between Leq and L95

The average difference between nighttime Leq and L95 is 8.4dB(A) and is close to legitimate limit. The disadvantages of the legal standard may still exist at nighttime and even subjectively more apparent because of the quiet environment at night.

As Figure 6 shows, No.47 and 65 are the two regions whose Leq and L95 differences exceed 15dB(A). When the difference exceeds 15dB(A), especially at nighttime, it may cause sleep disturbance. There are 16 cells whose Leq and L95 difference are greater than 10dB(A). This should raise enough concerns to prevent further deteriorating of the noise environment.

2.2.7. Noise Level Differences Associate with Cell Function

In order to evaluate the data more comprehensible and thoroughly, the data were re-classified by different functions of each cell.

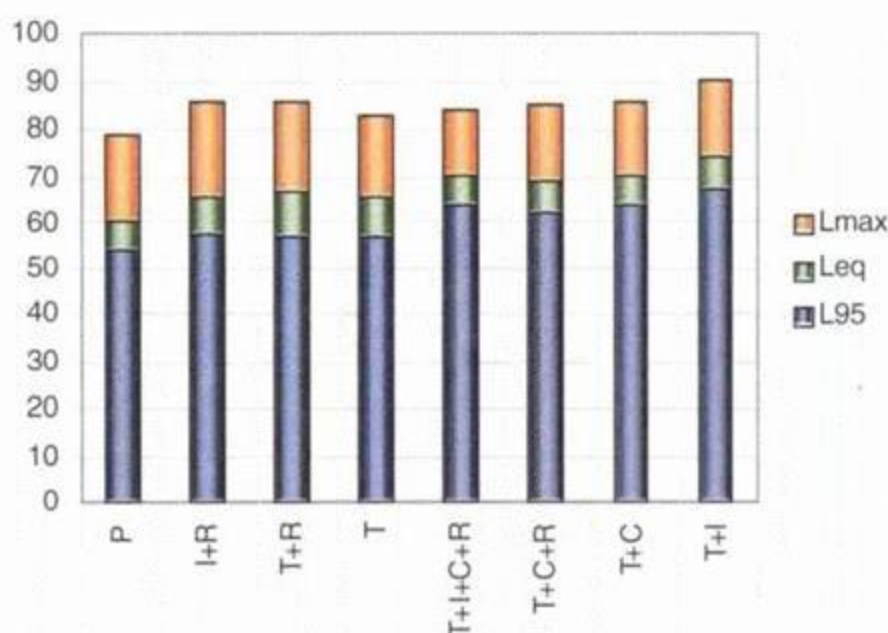
As Figure 7 and Figure 8 show, park is the quietest type of all the functions both at day and nighttime. Traffic and industrial combination is the noisiest. Similarly, traffic and commercial is the second noisiest combination. If we use the P.R.C. standard of roadside noise level as criterion, visibly, these two types can fulfill the standard neither at day nor nighttime. Besides these two types, the combination of traffic, industrial, commercial and residential noise also goes beyond the standard.

These data prove that traffic is the main noise source in Macao peninsular. Moreover, industrial and commercial noise sources are also important factors that affect the community's noise environment.

3. Regular Environmental Noise Monitoring

As the acoustical environment deteriorated with the city development, it is essential to set up a regular monitoring program to understand the noise environment in Macao. For this reason, in 1997, 12 stations of regular monitoring were set up in Macao peninsular. These 12 stations were selected according to different land use. In 1999, in order to profound the knowledge of

acoustic situation of Macao peninsular, another 8 stations were added into the regular environmental noise monitoring. These 20 stations cover most area of Macao peninsular.



(C-Commercial, I-Industrial, P-Park, R-Residential)

Figure 7 Daytime Noise Level of Different Cell Functions /dB(A)

3.1. Data Analysis Overall Noise Situation

Only the data of 1999 and 2000 are selected to evaluate the overall noise situation in Macao peninsular. *Figure 9* shows the average daytime noise situation at those 20 stations. The horizontal blue line represents P.R.C. noise standard for daytime roadside noise limit, and the red line is the legitimate reference background noise level in Macao peninsular.

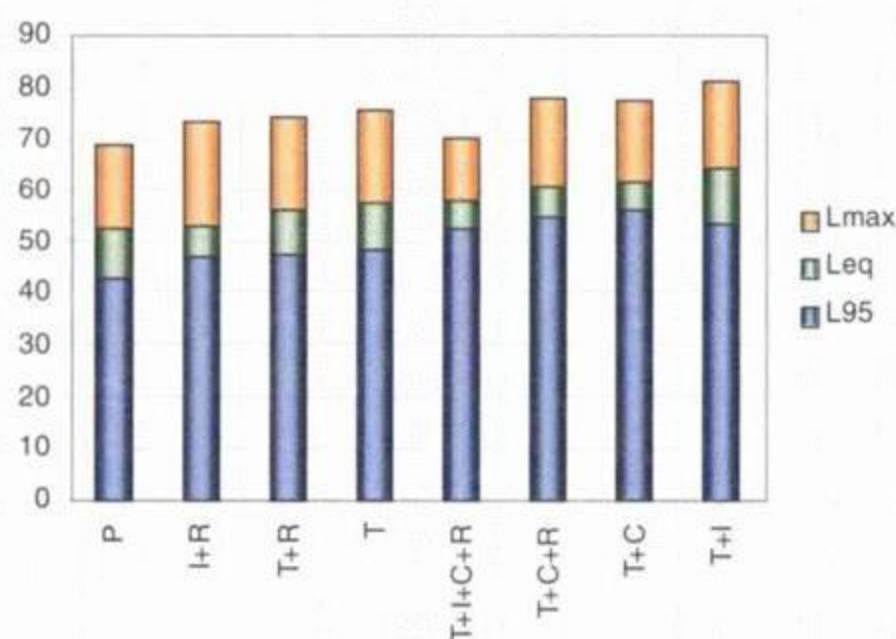
3.1.1. Leq

The average equivalent noise level of 20 stations is 71.4dB(A) and slightly exceeds the P.R.C. roadside limit. Only 4 stations' Leq values are less than 70dB because they are located at places where has less traffic noise.

On the other hand, the equivalent noise level of 3 stations (No.11, 8 and 7) is 5dB(A) higher than the P.R.C. stand. Obviously, these stations represent the noisiest area in Macao peninsular. Though these 3 stations are located in different region of Macao, they are all set on the busy roadside. The traffic volume of Station 11 is around 4000 vehicles per hour, while Station 8 and 7 are around 3000 veh/hr. Clearly, the environmental noise levels of these stations are closely related to the traffic volume. The larger the traffic flow is, the higher the noise level is. Besides the close relationship between the quantity of traffic and noise level, vehicle types also play an important role in environmental noise. This can be seen that the heavy-duty vehicles produce much higher level of noise than light duty vehicles. Station 11 is the noisiest, not only because of the very high traffic volume, but also because of having more than 800 heavy-duty vehicles per hour.

3.1.2. L95

The L95 value of a station is also influence by the types and intensity of human activities. Industrial regions, commercial centers or large traffic volume areas are likely to have higher background noise level. Station 11, 8, 7, 15 and 5 are the examples for large traffic volume while station 1 is the example for industrial region.



(C-Commercial, I-Industrial, P-Park, R-Residential)

Figure 8 Nighttime Noise Level of Different Cell Functions /dB(A)

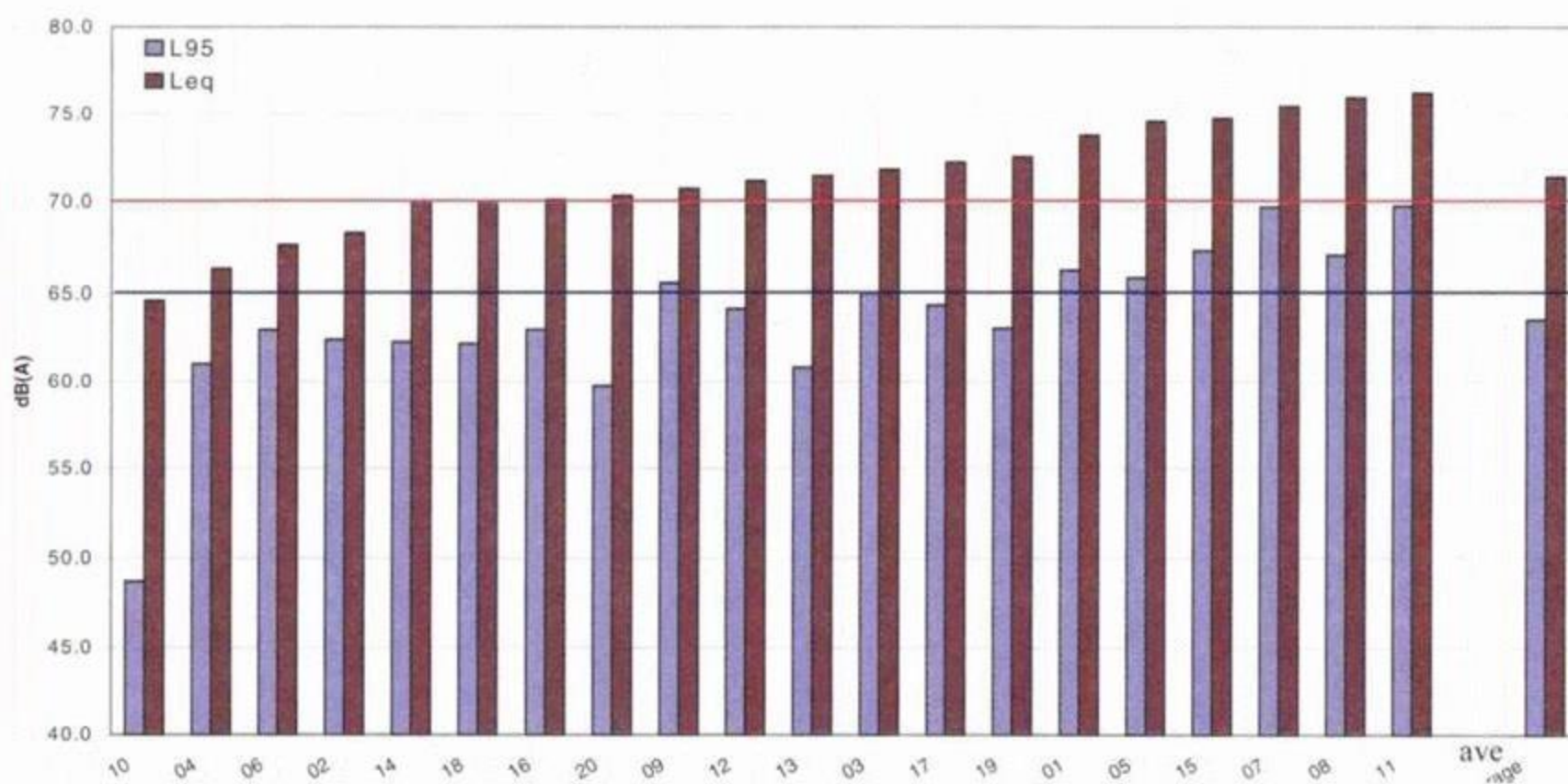


Figure 9 Daytime Leq and L95 of 20 Regular Monitoring Stations

3.2. Yearly Variations of Noise environment

Figure 10 shows the year average Leq data of 12 stations from 1997 to 2000. The 1998 data was special. Generally, the Leq data dropped in 1999. This result happened to match with the economic recession after the financial crisis in 1998. Station 10 appeared to be the most sensitive area to economic change. After the crisis, tourists from Hong Kong, Japan, Taiwan and other countries declined a lot. Fewer visitors mean less tourist buses. Because tourist buses are the major noise source of site 10, the Leq fell with decrease of buses. (Figure 11)

However, the background noise environment has less connection with the economic situation. The background noise level of each site was comparably steady for all the monitoring years.

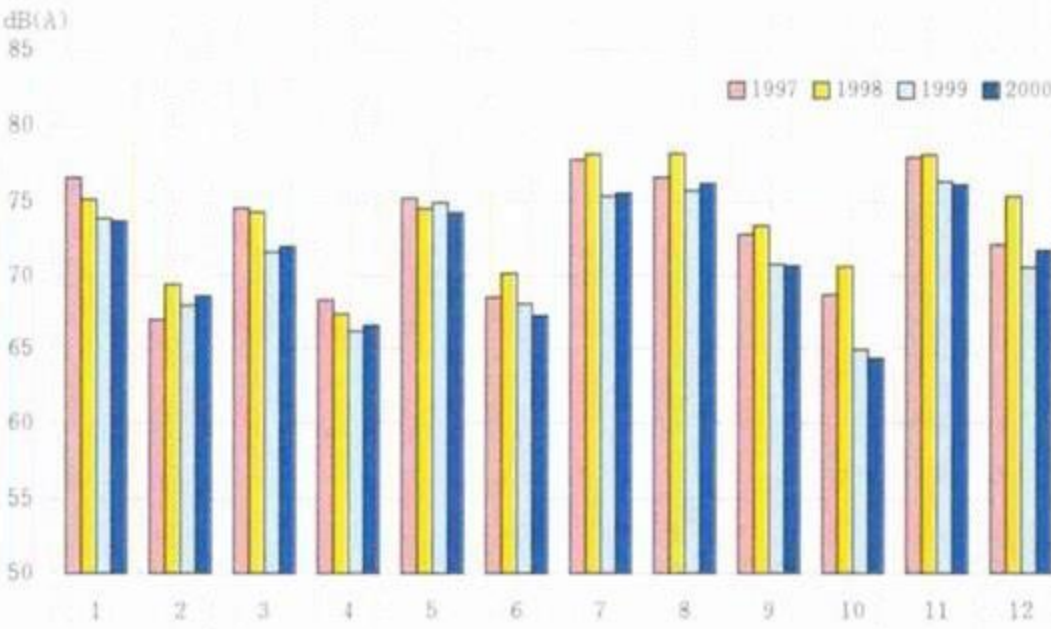


Figure 10 The Comparison of Year Average Leq

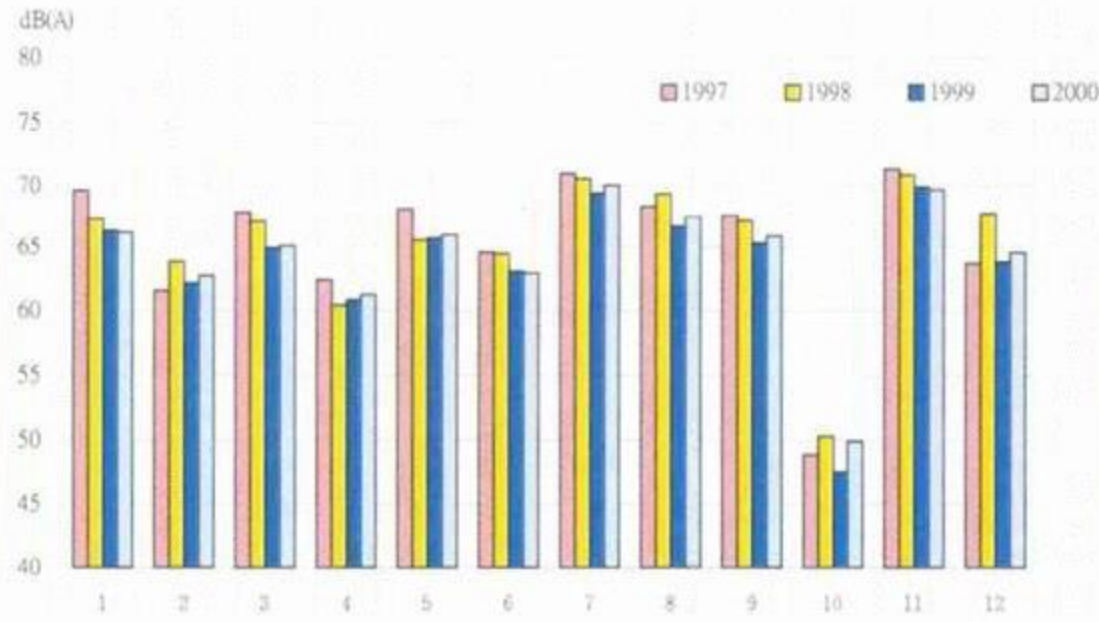


Figure 11 The Comparison of Year Average L95

4. Traffic Noise Simulation

As traffic is the most important noise source in Macao peninsular, it deserves to conduct particular study and analysis on this issue. Every single vehicle that runs on the roads or streets does its contribution to the noise environment. Since each type of vehicles produce a relatively certain level of noise, using traffic volume as a key parameter to simulate traffic noise is commonly used worldwide. In this study, STAMSON, which is a model developed in Canada, was employed to simulate and to predict the traffic noise level in Macao peninsular after adjusting some parameters.

4.1. Current Environmental Noise Simulation

The amount and average speed of different types of vehicles are vital to traffic noise prediction. According to the actual traffic situation in Macao peninsular, Table 1 lists the referenced noise level of different types of vehicles.



Figure 12 Traffic flow of survey roads in Macao peninsular

Table 1 Referenced Noise Level

original vehicle types	AU	MC	BU	LT	MT	HT
noise level /dB	65.0	71.0	71.0	67.5	72.5	74.6
the ratio to AU	1.0	1.09	1.09	1.04	1.12	1.14
STAMSON category	AU		MT		HT	

Notes: AU-- automobile; LT-- light truck; MT-- medium truck;
HT-- heavy truck; MC-- motorcycles; BU--bus.

Figure12 shows that, the total traffic flows of *Rua da Ribeira do Patane*, *Avenida do Almirante Lacerda* and *Avenida de Almeida Ribeiro* are very busy, and these roads have comparatively high level of noise.

Not only does the traffic flow affect the noise level, but also the speed of vehicles does. In this research, according to the situation in Macao, the vehicular speed is divided into three parts: 0 to 20 km/hr, 20 to 40 km/hr, and over 40 km/hr. Since the vehicular speed on many roads do not exceed 40km/hr, 40km/hr is selected as the maximum speed while using STAMSON to do the simulation.

In addition, because many roads in Macao have some gradient, and because the gradient of a road has some effects on the running situation of a vehicle and the noise it produces, the gradient of each researched road has been investigated. For simplicity, and due to the less importance of this parameter, the gradient of each road is classified as 0 to 5 degree, 5 to 15 degree, and 15 to 30 degree.

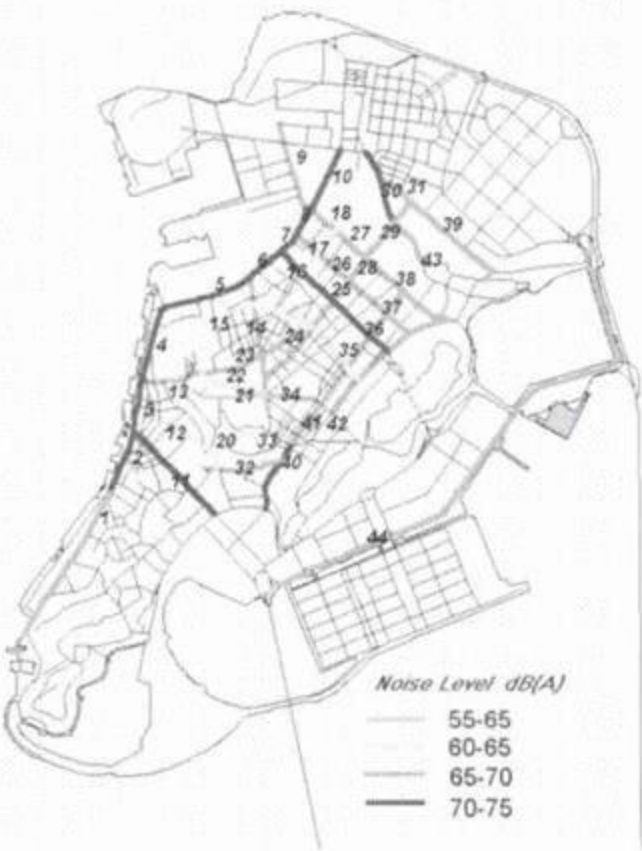


Figure12 Leq simulation results of survey roads in Macao peninsular

Simulation Result

The noise generated by vehicles, either because of its level or because of its impact to the surroundings, is the most important noise source in modern cities. The intensity of traffic noise, while other factors are comparably unvarying, greatly depends on the traffic flow and the speed of vehicles, also gradient of the road has some effects as well.

The simulation results have to compare with the regular monitoring data to examine their accuracy. The regular monitoring data of year 1999 are used because the traffic flow survey was done in that year. However, since not all of the regular monitoring sites were covered in the area of traffic survey, only the data of 8 stations are compared.

For the site located at roadside, the simulation result is the same as that road. For that located at a corner of the cross of two roads, the simulation result is the addition of the results of these two roads. The simulation results are showed in Table 2.

Table 2 Comparison between simulation results and monitoring data

Site No.		1	3	5*	7	8*	11*	12*	18*
simulation result	D1	73.4	70.2	74.8	74.4	74.6	75.7	70.3	70.6
	D2	74.4	71.3	73.6	74.4	76.2	77.3	71.8	71.1
	D3	72.7	69.6	74.5	73.7	74.0	75.3	69.9	71.8
	D4	73.9	70.7	74.9	74.8	75.1	76.2	70.8	69.7
	N1	71.1	67.5	73.4	72.0	71.3	73.5	66.7	68.0
	N2	66.4	63.2	68.6	67.6	68.5	68.6	64.4	59.7
	D-average	73.6	70.5	74.5	74.3	75.0	76.1	70.7	70.8
monitoring data	D1	73.6	71.3	74.8	75.0	75.1	76.2	70.2	70.2
	D2	74.3	71.3	74.8	75.1	75.7	76.5	70.1	69.4
	D3	73.9	70.4	74.3	75.3	75.5	76.2	70.4	69.6
	D4	73.9	72.1	75.1	75.6	75.6	76.4	70.8	70.4

	N1	69.8	68.4	73.4	73.5	73.4	75.0	69.0	66.5
	N2	63.2	62.6	58.6	68.0	68.8	67.8	63.2	62.5
	D-average	73.9	71.3	74.8	75.3	75.5	76.3	70.4	69.9

* -- The site located at the corner of a cross.

It can be seen that the simulation results matched with the monitoring data. The average errors are less than 2dB and are in the permissive range. The daytime simulation results cope with monitoring data even better. The less accuracy of nighttime simulation may be due to less precision of the nighttime traffic situation, while the traffic survey was mainly done at daytime.

5. Public Opinions on Noise

Since the annoyance of noise is subjective, the same level of noise may have very different response in different areas and at different period of time. In environmental noise legislation, these differences should take into account for better protecting the public health and welfare from the deleterious effects of noise.

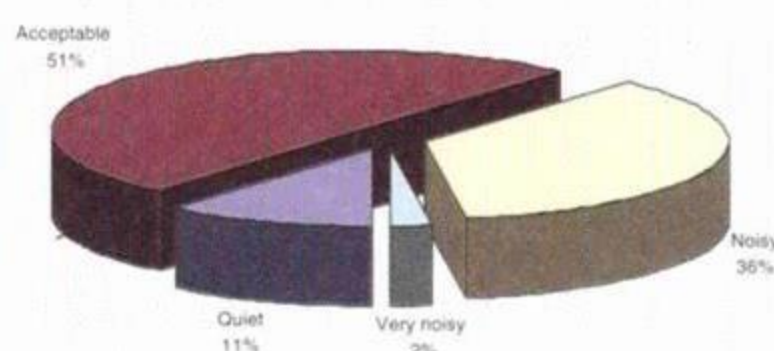


Figure 13 The overall impressions of noise environmental in Macao peninsular

5.1. Overall impression

In Macao peninsular, 51% of all interviewees rated the noise environmental as acceptable and 11% thought Macao is a quiet city. Among them, 36% cited that Macao is noisy, and only 2% of them regarded it as very noisy. (Figure 13)

Though the day and nighttime noise levels are high comparing with many other cities, the majority of the interviewees (more than 60%) considered the noise environment in Macao as acceptable or quiet. This illustrated that many Macao residents accustomed the situation and did not feel it much disturbing. On the other hand, the similarity of noise environment at most places in Macao peninsular, and the evenness of the noise level may also be the reasons of this result.

5.2. Neighborhood noise environment

At daytime, 15% of all the interviewees classify their neighborhood as quiet, 57% as acceptable and 28% as noisy. The nighttime results are similar to daytime with 27% quiet, 50% acceptable and 23% noisy.

Comparing the data of overall impression to those of neighborhood, though they are associated to some extent, these people's impressions towards the noise environment of their neighborhood are better than that of Macao peninsular. Because many Macao residents lived in high-storey-buildings, the distance of their apartments from the ground degraded the strength of noise from the streets.

At nighttime, about 77% interviewees cited that their neighborhoods are quiet or acceptable, more than that of daytime (5%). More people consider their neighborhoods are quiet at nighttime because many of the daytime activities stopped, fewer vehicles running in the city, most people off duty.

5.3. Noise disturbance

63% of all the interviewees reported that noise disturbed their daily life, mostly sleep. Among the disturbed people, 40% of them claimed that it happened at daytime, 49% at nighttime, and 11% at both day and nighttime. These data revealed that the high level of noise has affected the living quality of many Macao residents.

5.4. Noise sources

At daytime, 66% of all the interviewees thought traffic noise are the major source of noise in their daily life. Commercial noise is the second significant noise source with a percentage of 26. Domestic noise generated by their neighbors is rated as 16%. Traffic noise is the most important noise source in Macao, and the noise from commercial activities and their neighbors are also important.

At nighttime, the percentage of traffic noise had a noticeable decrease from 66% to 52%. Much less vehicles running on the streets at nighttime may result the reduction. In addition, the percentage of interviewees who claimed construction noise or industrial noise is the source of noise also dropped a lot because most construction works are prohibited and only a few factories are still operating at night.

On the contrast, the rate of interviewees who reported commercial noise and domestic noise increased at nighttime. Especially at early nighttime, when most residents are off duty, the prim time for restaurants and karaoke began. The noise generated by commercial activities became more significant. At this period of time, most people went back home, and the probability of producing noise escalated. At nighttime, traffic noise is still the major source and noise from commercial and domestic are other eminent sources.

6. Conclusion and Suggestion

- ◆ Restricted by the inadequate land area, most regions in Macao peninsular are for residential use, and at the same time commercial or industrial use embed in them. Moreover, since the network of land transportation covers Macao peninsular very densely, traffic use is inseparable from other kinds of land use. The traffic-industrial used region is the noisiest, while park is the quietest kind of land use.
- ◆ The average equivalent noise level of Macao peninsular is about 70dB(A). However, the places that are located beside the main drains of city transportation had the Leq exceeding 75dB(A). The average L_{95} is around 63dB(A) and is approximate to the legitimate reference 65dB(A). However, the legitimate reference had some restriction in actual application because the background noise level of Macao varied a lot. To consider public interest, the reference level should categorize for various land use.
- ◆ The noise environment in Macao peninsular is much affected by the traffic situation. Areas that have undesirable noise level also have very busy traffic flow. To control traffic noise, city planning, especially the planning of land transportation plays an important role. Moreover, to regulate the noise emission standard of different kinds of vehicles will also be effective to quiet the noise environment.
- ◆ Since the simulation results matched the monitoring data, using STAMSON to simulate or to predict the noise environment, especially the traffic noise level, is proper.
- ◆ 62% of all interviewees rated the noise environmental of Macao peninsular as acceptable or quiet. 63% of all the respondents reported of noise annoyance. Traffic noise is the most easily perceptible and the major source of noise at both day and nighttime. Commercial and domestic noise of the neighborhood is the other important sources of noise.

